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Dedicated to the bright memory of Professor Vadim F. Krotov

Krotov method for optimal control of closed quantum systems

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Abstract. The mathematics of optimal control of quantum systems is of great interest in connection with fundamental problems of physics as well as with existing and prospective applications to quantum technologies. One important problem is the development of methods for constructing controls for quantum systems. One of the commonly used methods is the Krotov method, which was initially proposed outside of quantum control theory in articles by Krotov and Feldman (1978, 1983). This method was used to develop a novel approach to finding optimal controls for quantum systems in [64] (Tannor, Kazakov, and Orlov, 1992), [65] (Somlói, Kazakov, and Tannor, 1993), and in many other works by various scientists. Our survey discusses mathematical aspects of this method for optimal control of closed quantum systems. It outlines various modifications with different forms of the improvement function (for example, linear or linear-quadratic), different constraints on the control spectrum and on the admissible states of the quantum system, different regularisers, and so on. The survey describes applications of the Krotov method to controlling molecular dynamics and Bose–Einstein condensates, and to quantum gate generation. This method is compared with the GRAPE (GRadient Ascent Pulse Engineering) method, the CRAB (Chopped Random-Basis) method, and the Zhu–Rabitz and Maday–Turinici methods.

Bibliography: 158 titles.

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Contents

1. Introduction

Optimal control theory considers optimal control problems (OCPs) for dynamical systems described by ordinary differential equations (ODEs), partial differential equations (PDEs), and so on. The theory began to develop in the middle of the 20th century, starting from such fundamental results as the Pontryagin maximum principle developed by Pontryagin, Boltyansky, Gamkrelidze and Mishchenko (see [1]) and the Bellman optimality principle [2]. Optimal control theory is now one of the leading areas of mathematics, with numerous applications in flight dynamics, robotics, economics, quantum technologies, and so on.

One area of application for optimal control theory is quantum control: an advanced interdisciplinary direction devoted to the control of quantum systems, that is, of individual electrons, atoms, molecules, and photons. Control is implemented by shaped laser pulses, modulating electromagnetic radiation, using a non-equilibrium reservoir, or other effects. Modern technologies allow one to produce laser pulses of ultra-short duration on the order of femtoseconds (10^{-15} sec) and attoseconds (10^{-18} sec). The great interest in mathematical problems of quantum control is connected with progress in experiments on the manipulation of quantum systems. In 1997, the Nobel Prize in Physics was awarded to S. Chu, C. Cohen-Tannoudji, and W. Phillips "for development of methods to cool and trap atoms with laser light".[1] In 2001, the Nobel Prize in Physics was awarded to E. Cornell, W. Ketterle, and C. Wieman, who in 1995 made Bose–Einstein condensate in their laboratories.[2] In 2012, the Nobel Prize in Physics was awarded to S. Haroche and D. Wineland "for ground-breaking experimental methods that enable measuring and manipulation of individual quantum systems".[3] In 2018, the Nobel Prize in Physics was awarded "for ground-breaking inventions in the field of laser physics" to A. Ashkin "for the optical tweezers and their application to biological systems" and to G. Mourou and D. Strickland "for their method of generating high-intensity, ultra-short optical pulses".[4]

The mathematics of quantum control has been actively studied since the 1980s. Quantum control is important for existing and prospective technologies, including: control of atomic and molecular dynamics (for example, laser cooling of molecules); manipulation of Bose–Einstein condensate; implementation of quantum computing (for example, for optimal generation of qubits and quantum gates); design of atomic chips; laser-assisted isotope separation; laser chemistry; nuclear magnetic resonance; dynamic nuclear polarisation; magnetic resonance imaging, and so on. The theoretical and experimental results on quantum control are summarised in the books and surveys by: Butkovskiy and Samoilenko (1984) [3]; Krasnov, Shaparev, and Shkedov (1989) [4]; Rice and Zhao (2000) [5]; Bandrauk, Delfour, and Le Bris (editors,2003) [6]; D'Alessandro (2003) [7]; Shapiro and Brumer (2003) [8]; Tannor (2007) [9]; Letokhov (2007) [10]; D'Alessandro (2007) [11]; Fradkov (2007) [12]; Brif, Chakrabarti, and Rabitz (2010) [13]; Dong and Petersen (2010) [14]; Wiseman and Milburn (2010) [15]; Altafini and Ticozzi (2012) [16]; Bonnard and Sugny (2012) [17]; Gough (2012) [18]; Cong (2014) [19]; Dong, Wu, Yuan, Li, and Tarn

[1] https://www.nobelprize.org/prizes/physics/1997/summary/
[2] https://www.nobelprize.org/prizes/physics/2001/summary/
[3] https://www.nobelprize.org/prizes/physics/2012/summary/
[4] https://www.nobelprize.org/prizes/physics/2018/summary/

(2015) [20]; Glaser, Boscain, Calarco, Koch, Köckenberger, Kosloff, Kuprov, Luy, Schirmer, Schulte-Herbrüggen, Sugny, and Wilhelm (2015) [21]; Koch (2016) [22]; Borzì, Ciaramella, and Sprengel (2017) [23].

The optimal control of quantum systems includes *program* control, where the control function depends on the time, and *feedback* control, where the control function depends on the time and the measured system's state. The article [24] considers quantum control for discretely observable quantum systems which evolve between observations according to the Schrödinger equation. The article [25] contains the theory of real-time feedback control for physical models of quantum optics. For the construction of program quantum control, various methods are used: reduction to finite-dimensional optimisation followed by the use of probabilistic global search algorithms, for example, evolutionary genetic algorithms [26], [27] and dual annealing [28]; the Pontryagin maximum principle and geometric control (for example, for minimal-time quantum control) [29]–[37]; gradient flows [38]; GRAPE [39], [40] and CRAB [41]; the Zhu–Rabitz [42] and the Maday–Turinici [43] methods; dynamic programming [44], [45]; time-parallelised algorithms [46]; the speed-gradient method [47], [48]; the Ho–Rabitz TBQCP (Two-point Boundary-value Quantum Control Paradigm) method [49]; the gradient-projection method [28], [50]. Machine learning is also used in research on quantum systems and technologies: reinforcement learning is applied to construct quantum controls [51], [52]; auto-encoders are applied to reduce the dimensionality of data describing quantum dynamics [53]; the restricted Boltzmann machine is used for quantum tomography [54], [55]. Quantum machine learning is also considered [56].

One commonly used method for constructing program controls for quantum systems is the Krotov[5] method. This method was initially proposed outside of quantum control by Krotov and Feldman [57], [58] (1978, 1983) based on the Krotov optimality principle [59], [60] and was further developed by Konnov and Krotov [61] (1999). An example involving control of an open quantum system (that is, one interacting with the environment) was analysed by Kazakov and Krotov in 1987 [62] (see also [63]). A crucial step in its application to quantum control was made in 1992–1993, when Tannor and his coauthors used the first-order Krotov method to develop a general approach for finding optimal controls for quantum systems [64], [65]. In 2002, the second-order Krotov method [58], [61] was adapted by Sklarz and Tannor for optimal control of a Bose–Einstein condensate, whose dynamics is defined in terms of a controlled Gross–Pitaevskii equation [66]. In 2008, Palao, Kosloff, and Koch developed the method of optimal control in the problem of reaching a target in a certain subspace of a Hilbert space while avoiding population transfer to other subspaces [67]. With various modifications and

[5]Vadim Fedorovich Krotov (1932–2015) was a well-known researcher, an Honoured Scientist of the Russian Federation and an author of fundamental results in optimal control theory. In 1962 he defended his Ph.D. thesis "A new method of variational calculus and some of its applications" at the Steklov Mathematical Institute of the USSR Academy of Sciences, and in 1963 he defended his D.Sc. thesis "Some new methods of variational calculus and their application to flight dynamics" at the Moscow Aviation Institute. He was the head of departments in the Moscow Aviation Technological Institute and the Moscow Institute of Economics and Statistics. In 1982–2015 he worked in the Institute of Control Sciences of the Russian Academy of Sciences (ICS RAS) as the head of Laboratory 45, which now bears his name. The website of ICS RAS has a page devoted to V. F. Krotov: http://www.ipu.ru/node/32378 (in Russian).

with account taken of specific details of quantum optimal control problems, the Krotov method was applied to: manipulation of atomic and molecular dynamics ([9], [19], [43], [67]–[78]); generation of qubits states, quantum gates, quantum networks ([79]–[96]); manipulation of a Bose–Einstein condensate ([40], [66], [97], [98]); nuclear magnetic resonance, dynamic nuclear polarisation, and magnetic resonance imaging ([99]–[101]). The research group of Koch and her coauthors has developed program tools in Fortran and Python ([102], [103]) that include implementations of both the first- and the second-order Krotov methods and that also involve constraints on quantum states and the control spectrum.

There are many dissertations on quantum optimal control which use the iterative Krotov method. These include, for example: the dissertation [104] by Reich (2015) on the foundations of quantum optimal control for open quantum systems; the dissertation [79] by Goerz (2015) on the optimisation of robust quantum gates for open quantum systems; and the dissertation [97] by Jäger (2015) on optimal control of a Bose–Einstein condensate.

This survey outlines mathematical results and applications of the Krotov method to closed (that is, not interacting with the environment) quantum systems evolving under a coherent control in the perturbed part of the Hamiltonian. Open quantum systems will be considered elsewhere. The survey does not claim to be a complete overview of all modifications of the method for quantum systems, and only basic results are given. We use the term 'Krotov method' following the tradition established in quantum optimal control. At the same time, the publications [57] and [58] (Krotov and Feldman) and [61] (Konnov and Krotov) were in co-authorship, and this may be reflected in the name of the method.

The structure of the survey is as follows. Section 2 provides formulations of OCPs for closed quantum systems. It also includes a brief discussion of controllability and control landscapes for such systems. Section 3 is devoted to the Krotov method for OCPs with real-valued states. Section 4 discusses the first-order Krotov method and the Zhu–Rabitz and Madey–Turinici methods for systems governed by the Schrödinger equation and the Liouville–von Neumann equation. Section 5 considers the generation of target unitary transformations and control of ensembles of quantum states. Section 6 discusses applications of the Krotov and GRAPE methods to the manipulation of a Bose–Einstein condensate whose dynamics is governed by the controlled Gross–Pitaevskii equation. Section 7 (Conclusions) summarises the survey.

2. Classes of optimal control problems for closed quantum systems

In this section we consider formulations of optimal control problems for closed quantum systems. A formulation of an optimal control problem involves setting a dynamical equation, a space of controls, a cost functional to be minimised, and constraints on the controls and admissible states of the system.

Each quantum system is associated with some Hilbert space $\mathscr{H}$: for example, $\mathscr{H} = \mathbb{C}^n$ and $\mathscr{H} = L^2(\Omega;\mathbb{C})$, respectively, for a system with n states and for a quantum particle moving in a domain $\Omega \subseteq \mathbb{R}^d$. Pure states of the system are unit-norm vectors $\psi \in \mathscr{H}$, $\|\psi\|^2 = 1$. Some models consider the Hilbert space $L^2(\Omega;\mathbb{C}^M)$, $\Omega \subseteq \mathbb{R}^q$; a state is a vector function $\psi = (\psi_1, \dots, \psi_M)$. Most general

states of a quantum system are described by density matrices. A density matrix is a self-adjoint trace-class operator ρ acting in $\mathscr{H}$ which satisfies the conditions $\rho \geqslant 0$ and $\operatorname{Tr}\rho = 1$.

The dynamics of the system state in the absence of controls is determined by a free-system Hamiltonian $\mathbf{H}_0(t)$, which is a self-adjoint operator acting in $\mathscr{H}$. The norm of the state is a dynamical invariant: $\|\psi(t)\|^2_{\mathscr{H}} = 1$. In the case of an electron in $\Omega \subset \mathbb{R}^3$ this means that the probability of detecting an electron in the domain Ω at any time t is 1. For a qubit, $n = 2$ and $\psi(t) = \alpha(t)|0\rangle + \beta(t)|1\rangle \in \mathbb{C}^2$ (here $|0\rangle = \begin{pmatrix}1\\0\end{pmatrix}$ and $|1\rangle = \begin{pmatrix}0\\1\end{pmatrix}$ in the Dirac notation), and $|\alpha(t)|^2 + |\beta(t)|^2 = 1$, which implies that $\|\psi(t)\|^2_{\mathbb{C}^2} = 1$. The value of $|\alpha(t)|^2$ is the probability of finding the system in the pure state $|0\rangle$, and the value of $|\beta(t)|^2$ is the probability of finding the system in the system in the pure state $|1\rangle$. We consider m controls, so that the interaction with the control u_l $(l = 1, \ldots, m)$ is described by a self-adjoint interaction Hamiltonian $\mathbf{H}_l$. An observable O of the system is a self-adjoint operator acting in $\mathscr{H}$. If the system is in a pure state ψ, then the mean observed value of O is $\langle O\rangle = \langle \psi, O\psi\rangle$. If the system is in the state with density matrix ρ, then $\langle O\rangle = \operatorname{Tr}(\rho O)$.

2.1. Schrödinger equation with controlled Hamiltonian and cost criteria. Based on [3], [11], and [23], in particular, we formulate the following definition.

Definition 2.1. A system with Hilbert space $\mathscr{H}$ and governed by the Schrödinger equation with a linearly controlled Hamiltonian $\mathbf{H}$ is a quantum system whose state $\psi(t) \in \mathscr{H}$ satisfies the equation

$$\frac{d\psi(t)}{dt} = -\frac{i}{\hbar}\mathbf{H}[u(t)]\psi(t), \qquad \psi(0) = \psi_0, \tag{2.1}$$

where

$$\mathbf{H}[u(t)] = \mathbf{H}_0 + \sum_{l=1}^{m} \mathbf{H}_l u_l(t), \tag{2.2}$$

$$u \in \mathscr{U} = \mathrm{PC}([0,T]; Q), \qquad Q \subseteq \mathbb{R}^m. \tag{2.3}$$

The initial state $\psi_0 \in \mathscr{H}$ and the final moment of time T are fixed; $\hbar$ is the Planck constant; $u(t) = (u_l(t))_{l=1,\ldots,m}$ is a vector control function; $\mathscr{U}$ is the class of admissible controls; Q is a convex set; the Hamiltonian $\mathbf{H}$ is a self-adjoint operator acting in the Hilbert space $\mathscr{H}$; the operator $\mathbf{H}_0$ is the unperturbed part of $\mathbf{H}$; the operator $\mathbf{H}_l$ characterises the interaction of the quantum system with the external control $u_l(t)$.

As the class of admissible controls $\mathscr{U}$, we will consider the space $\mathrm{PC}([0,T]; Q)$ of piecewise continuous functions. The time T can be of the order of femtoseconds, picoseconds, and so on.

The following theorem is a corollary of Carathéodory's theorem on the existence and uniqueness of the solution of a differential equation with discontinuous right-hand side.

Theorem 2.1. *Let $\mathscr{H} = \mathbb{C}^n$. If $u \in L^1([0,T];Q)$, then the solution of (2.1) exists in the class of absolutely continuous functions on the interval $[0,T]$ and is unique.*

Since $\mathrm{PC}([0,T];Q) \subset L^1([0,T];Q)$, for $\mathscr{H} = \mathbb{C}^n$ the solution of (2.1) exists and is unique for each piecewise continuous control u.

Definition 2.2. The process $v = (\psi(t), u(t) \mid t \in [0,T])$ is said to be admissible if it satisfies the conditions (2.1)–(2.3).

We denote by $\mathscr{D}$ the set of all admissible processes.

Remark 2.1. In the general case the control u can be a complex-valued function ([9], [64], [105]). In what follows we consider real-valued controls as specified in (2.3). In the general case, the Hamiltonian $\mathbf{H}$ can depend non-linearly on the control u [106]. Below we consider only the linear case, because it is commonly used.

Definition 2.3. For the system (2.1)–(2.3), the following OCP is referred to as the problem of maximising the mean $\langle O \rangle = \langle \psi(T), O\psi(T) \rangle$ for a Hermitian operator O:

$$J(v) = \mathscr{F}_O(\psi(T)) + \lambda_u \int_0^T \frac{\|u(t)\|^2}{S(t)}\, dt + \lambda_\psi \int_0^T \langle \psi(t), D(t)\psi(t) \rangle\, dt \to \min_{v \in \mathscr{D}}, \qquad J\colon \mathscr{D} \to \mathbb{R}, \tag{2.4}$$

where the cost criterion contains the terminant $\mathscr{F}_O = -\langle O \rangle$ and the parameters $\lambda_u \geqslant 0$ and $\lambda_\psi \leqslant 0$, the operator $D(t)$ ($t \in [0,T]$) is self-adjoint and positive-semidefinite, and S is some shape function.

For the first term on the right-hand side of (2.4) we can consider the condition $O \geqslant 0$, which is essential for successive improvements of controls for OCPs of the type (2.1)–(2.4) with $\lambda_\psi = 0$ using the methods given in [64] (Tannor, Kazakov, and Orlov, 1992), [42] (Zhu and Rabitz, 1998), and [43] (Maday and Turinici, 2003).

The second term on the right-hand side of (2.4) can be regarded as a condition for energy minimisation, which is important for avoiding non-physical coherent-control values, or as a possibility for simplifying the application of optimisation methods. This term can change the original OCP significantly by affecting the value of the terminant $\mathscr{F}(\psi(T))$, which is why adjustment of λ_u is necessary. Along with $S(t) \equiv 1$ ([42], [64], [65], [107], [108]), a non-constant function S can be used to ensure a smooth turn-on of the laser field at time $t = 0$ and a smooth turn-off at time $t = T$. Examples include $S(t) = \sin^2(\pi t/T)$ (Sundermann and de Vivie-Riedle [109], 1999), $S(t) = \exp[-32(t/T - 1/2)^2]$ (Palao, Kosloff, and Koch [67], p. 5, 2008), and so on.

The third term with the operator $D(t)$ allows one to specify forbidden or allowed subspaces of $\mathscr{H}$ [67]. If the operator $D(t)$ is positive-semidefinite, then it describes an allowed subspace. The role of this term was discussed in [67] in detail. The requirement $\lambda_\psi \leqslant 0$ is chosen by taking into account a special condition for a non-decreasing cost functional using the first-order Krotov method considered in Section 4.

Constraints of the type

$$\int_0^T u^2(t)\, dt\ \{=, \leqslant\}\ E$$

(E is some given value) or, in the complex-valued case, of the type

$$\int_0^T u(t)u^*(t)\,dt\ \{=,\leqslant\}\ E$$

(see [62], [105], [108]) can also be used. In this survey we do not consider such constraints.

Along with $\mathscr{F}_O$, one considers terminants with a given target state $\psi_{\text{target}} \in \mathscr{H}$ (see, for example, [3] and [110]):

$$\mathscr{F}_{\psi_{\text{target}}} = 1 - |\langle \psi(T), \psi_{\text{target}} \rangle|^2 \tag{2.5}$$

and

$$\mathscr{F}_{\psi_{\text{target}}} = 1 - \operatorname{Re}\langle \psi(T), \psi_{\text{target}} \rangle = \frac{1}{2}\|\psi(T) - \psi_{\text{target}}\|^2. \tag{2.6}$$

Minimising (2.5) means maximising the probability that the final state $\psi(T) \in \mathscr{H}$ is the target state ψ_{target}.

With respect to $\mathscr{F}_O(\psi(T))$ in (2.4), the terminant (2.5) represents a particular case, where $O = P_{\text{target}}$ is the projection on the target state ψ_{target}. For $M = 1$ and $x \in \mathbb{R}^3$ one can, for example, take ψ_{target} to be a sum of Gaussian functions (see [74], p. 6):

$$\psi_{\text{target}}(x) = A\left[\exp\left(-\sum_{j=1}^{3} \alpha_j (x_j - x_j^{\alpha})^2\right) + \exp\left(-\sum_{j=1}^{3} \beta_j (x_j - x_j^{\beta})^2\right)\right],$$

with $\alpha_j, \beta_j > 0$. The following term (see [110], p. 4), which sets a condition on the trajectory over the entire time range, is sometimes also considered in the cost functional J:

$$\mathscr{F}_{\psi_{\text{target}}(t)} = \frac{1}{2}\int_0^T \|\psi(t) - \psi_{\text{target}}(t)\|^2\,dt = T - \int_0^T \operatorname{Re}\langle \psi_{\text{target}}(t), \psi(t)\rangle\,dt.$$

Definition 2.4. For the OCP (2.1)–(2.4), an admissible process v^* is a solution if this process provides the global minimum $J(v^*) = \min_{v\in\mathscr{D}} J(v)$ of the cost functional J.

In some cases an OCP for the Schrödinger equation in an infinite-dimensional Hilbert space can be reduced to an approximate OCP for a finite-dimensional system, and for the latter OCP one can consider the corresponding OCP with real-valued states. The articles [77] and [111] consider the equation

$$i\frac{\partial \psi(\theta, t)}{\partial t} = \left(-\frac{\partial^2}{\partial \theta^2} + u(t)\cos\theta\right)\psi(\theta, t), \qquad \psi(0) = \psi_0, \quad \theta \in \Omega,$$

describing rotation of a planar molecule, and the following approximate OCP:

$$\frac{dz(t)}{dt} = \big(A + u(t)B\big)z(t), \qquad z(0) = z_0, \qquad z(t) \in \mathbb{C}^n, \tag{2.7}$$

$$J(z, u) = |z_2(T)|^2 \to \min. \tag{2.8}$$

Here θ is the angle between the polarisation direction and the molecular axis, Ω is a one-dimensional torus, A and B are $n \times n$ matrices obtained in approximation by the Galerkin method, and $n = 22$.

2.2. Liouville–von Neumann equation with control and cost criteria. The evolution of the density matrix of a closed quantum system under a control is described by the Liouville–von Neumann equation with controlled Hamiltonian:

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}\big[\mathbf{H}[u(t)], \rho\big], \qquad \rho(0) = \rho_0, \tag{2.9}$$

where $[\,\cdot\,,\cdot\,]$ is the commutator ($[A, B] = AB - BA$) and the initial density matrix ρ_0 is given.

If a quantum system is open, that is, is interacting with the environment (a reservoir), then the evolution of the density matrix $\rho(t)$ under the influence of a coherent control will not be unitary and can be described by the equation

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}\big[\mathbf{H}[u(t)], \rho\big] + \mathscr{L}(\rho), \qquad \rho(0) = \rho_0, \tag{2.10}$$

where the dissipator $\mathscr{L}(\rho)$ can have, for example, the Gorini–Kossakowski–Sudarshan–Lindblad form

$$\mathscr{L}(\rho) = \sum_k \gamma_k \left(A_k \rho A_k^\dagger - \frac{1}{2}\{A_k^\dagger A_k, \rho\}\right).$$

Here the A_k are the Lindblad operators which model different dissipation channels, $\{\,\cdot\,,\cdot\,\}$ is the anticommutator ($\{A, B\} = AB + BA$), and the $\gamma_k \geqslant 0$ are parameters. If $\mathscr{L}(\rho) \equiv 0$, then the equation (2.10) becomes the Liouville–von Neumann equation (2.9).

Definition 2.5. For the system (2.9), the problem of maximisation of the mean

$$\langle \rho_{\text{target}} \rangle = \operatorname{Tr}\big(\rho(T)\rho_{\text{target}}\big)$$

(see [3] and [110]) among the final density matrix $\rho(T)$ and the target density matrix ρ_{target} is defined by the following condition:

$$J(v) = \mathscr{F}_{\rho_{\text{target}}}(\rho(T)) + \lambda_u \int_0^T \frac{\|u(t)\|^2}{S(t)}\, dt + \lambda_\rho \int_0^T \operatorname{Tr}\big(\rho(t)D(t)\big)\, dt \to \min_{v \in \mathscr{D}}, \tag{2.11}$$

where the terminant $\mathscr{F}_{\rho_{\text{target}}}$ is equal to $-\langle \rho_{\text{target}} \rangle$, $v = (\rho, u)$, $\lambda_u \geqslant 0$ and $\lambda_\rho \leqslant 0$ are parameters, and the operator $D(t)$ is self-adjoint and positive-semidefinite.

2.3. Cost criteria for unitary transformations and for the ensemble of solutions of the Schrödinger equation. Potential applications of quantum technologies include quantum computations, which could significantly increase the speed of solution of complex problems such as factorising a large number or searching in an unsorted database [112]–[115]. Basic objects in quantum computation include the *qubit* (quantum bit), which is a two-state quantum system, and the *quantum gate*, which is an elementary operation transforming input states of one or more qubits into certain output states.

Mathematically, a quantum gate is a unitary matrix W (see the monographs [115]–[117]). For example, the Hadamard gate is the following one-qubit ($n = 2$) unitary matrix:

$$W_{\text{H}} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix}.$$

Two-qubit gates ($n = 2^2$) include, for example, gates such as the controlled NOT (CNOT), the quantum Fourier transform (QFT), the controlled phase gate (CPHASE), and the BGATE:

$$W_{\mathrm{CNOT}} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{pmatrix}, \qquad W_{\mathrm{QFT}} = \frac{1}{2}\begin{pmatrix} 1 & 1 & 1 & 1 \\ 1 & i & -1 & -i \\ 1 & -1 & 1 & -1 \\ 1 & -i & -1 & i \end{pmatrix},$$

$$W_{\mathrm{CPHASE}} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix}, \quad W_{\mathrm{BGATE}} = \begin{pmatrix} \cos\frac{\pi}{8} & 0 & 0 & i\sin\frac{\pi}{8} \\ 0 & \cos\frac{3\pi}{8} & i\sin\frac{3\pi}{8} & 0 \\ 0 & i\sin\frac{3\pi}{8} & \cos\frac{3\pi}{8} & 0 \\ i\sin\frac{\pi}{8} & 0 & 0 & \cos\frac{\pi}{8} \end{pmatrix}.$$

Examples of three-qubit gates are the Toffoli gate and the Fredkin gate.

Implementation of a quantum gate W in a quantum processor means realisation of a suitable controlled physical process (see, for example, [80] and [81]) which produces the evolution of the system to the unitary matrix W.

Definition 2.6. The operator $U(t)$ satisfying the Cauchy problem

$$\frac{dU(t)}{dt} = -\frac{i}{\hbar}\Big(\mathbf{H}_0 + \sum_{l=1}^{m} \mathbf{H}_l u_l(t)\Big)U(t), \qquad U(0) = \mathbb{I}, \tag{2.12}$$

is called the unitary evolution operator of a quantum system with Hilbert space $\mathscr{H}$ and with control $u(t) = (u_1(t), \dots, u_m(t))$, where $\mathbf{H}_l$ $(l = 0, \dots, m)$ are Hermitian operators and $\mathbb{I}$ is the identity operator.

In analogy to Theorem 2.1 (see also Lemma 2.1 in [118], p. 315), the solution of (2.12) exists, is an absolutely continuous matrix function, and is unique for controls in $L^1([0,T]; Q)$.

We have $\psi(t) = U(t)\psi_0$. The evolution of the density matrix is given by

$$\rho(t) = U(t)\rho_0 U^\dagger(t). \tag{2.13}$$

The density matrix $\rho(t)$ satisfies the Liouville–von Neumann equation (2.9). From (2.13) it follows that

$$\frac{1}{2}\|\rho(T) - \rho_{\mathrm{target}}\|^2 = C - \mathrm{Tr}(\rho_{\mathrm{target}}\,\rho(T)),$$

where $C = \frac{1}{2}\,\mathrm{Tr}\,\rho_0^2 + \frac{1}{2}\,\mathrm{Tr}\,\rho_{\mathrm{target}}^2$. For trajectory optimisation the following functional is used (see [110], p. 4):

$$\frac{1}{2}\int_0^T \|\rho(t) - \rho_{\mathrm{target}}(t)\|^2\,dt = CT - \int_0^T \mathrm{Tr}\big(\rho_{\mathrm{target}}(t)\rho(t)\big)\,dt.$$

Definition 2.7. The OCP for the unitary operator $U(t)$ is the problem of minimising the cost functional

$$J_X(v) = \mathscr{F}_X(U(T)) + \lambda_u \int_0^T \frac{\|u(t)\|^2}{S(t)}\,dt, \qquad \lambda_u \geqslant 0, \quad v = (U, \rho), \tag{2.14}$$

for the system (2.12), where the terminant $\mathscr{F}_X$, $X \in \{W, O, A\}$, is defined in one of the following ways:

$$\mathscr{F}_W(U(T)) = -\frac{1}{n^2}|\operatorname{Tr}(W^\dagger U(T))|^2, \tag{2.15}$$

$$\mathscr{F}_O(U(T)) = -\operatorname{Tr}(O\rho(T)) = -\operatorname{Tr}(OU(T)\rho_0 U^\dagger(T)), \tag{2.16}$$

$$\mathscr{F}_A(U(T)) = -\operatorname{Re}[\operatorname{Tr}(A^\dagger\rho(T))] = -\operatorname{Re}[\operatorname{Tr}(A^\dagger U(T)\rho_0 U^\dagger(T))], \tag{2.17}$$

$$\mathscr{F}_A(U(T)) = -|\operatorname{Tr}(A^\dagger\rho(T))|^2 = -|\operatorname{Tr}(A^\dagger U(T)\rho_0 U^\dagger(T))|^2; \tag{2.18}$$

here W is unitary, O is self-adjoint, A is a non-self-adjoint operator, and S is some shape function.

Problems of this kind are considered, for example, in connection with nuclear magnetic resonance, magnetic resonance imaging, dynamic nuclear polarisation [99], [100], and quantum gate generation (see [119] and [120], for example).

The definition below follows [67], [76], [81], [86], [88]–[91], and [104].

Definition 2.8. The ensemble of solutions of the Schrödinger equation is a set of states $\{\psi_j(t) \mid t \in [0,T],\ j = 1,\dots,n\}$ whose jth element is the solution of the Schrödinger equation (2.1) with controlled Hamiltonian (2.2), where $\psi = \psi_j$, $\psi_0 = \psi_{j,0}$, the initial states $\{\psi_{j,0} \mid j = 1,\dots,n\}$ are prescribed, and the control u is the same for all $j = 1,\dots,n$.

Instead of a unitary transformation $U(t)$ we can consider the corresponding ensemble of solutions of the Schrödinger equation, because

$$\psi_j(T) = U(T)\psi_{0,j}, \quad \psi_{\text{target},j} = W\psi_{0,j} \quad \text{and} \quad j = 1,\dots,n,$$

where W is a target unitary transformation. For the ensemble of solutions, one can consider the corresponding OCP for controlled simultaneous transitions of the system from the set of initial states $\psi_{0,j}$ to the set of target states $\psi_{\text{target},j}$, $j = 1,\dots,n$.

Definition 2.9. The following problem is called an OCP for an ensemble of solutions of the Schrödinger equation:

$$\frac{d\psi_j(t)}{dt} = -\frac{i}{\hbar}\mathbf{H}[u(t)]\psi_j(t), \qquad \psi_j(0) = \psi_{0,j}, \quad j = 1,\dots,n, \tag{2.19}$$

$$J(v) = \mathscr{F}\big(\{\psi_j(T)\}_{j=1,\dots,n}\big) + \lambda_\psi \int_0^T \sum_{j=1}^n \langle\psi_j(t), D(t)\psi_j(t)\rangle\,dt \to \min, \tag{2.20}$$

where $v = \big(\{\psi_j\}_{j=1,\dots,n}, u\big)$ is the control process, the terminant $\mathscr{F}$ is defined on the set of final states of the ensemble of solutions, $\lambda_\psi \leqslant 0$ is a parameter, and the operator $D(t)$ $(t \in [0,T])$ is self-adjoint and positive-semidefinite.

The specific details of an OCP (2.19), (2.20) may differ ([67], [76], [81], [88]–[91], [104]). The articles [80], [81] (Palao and Kosloff, 2002, 2003) consider control of lithium and sodium molecules with two electronic states (ground and excited). The first $n = 2^q$ levels of the ground state are regarded as registers for q qubits, and the goal is to find a control u which provides the realisation of the target gate W. The article [80] considers OCPs for implementing the quantum gates W_{H} and W_{QFT} by realising simultaneous transitions between electronic surfaces for solutions ψ_j of the Schrödinger equation. The goal is to minimise the terminant

$$\mathscr{F}(U(T)) = -\frac{1}{n^2}|\operatorname{Tr}\{W^\dagger U(T) P_n\}|^2 = -\frac{1}{n^2}\sum_{j=1}^{n}\sum_{j'=1}^{n}\langle j|W^\dagger U(T)|j\rangle\langle j'|U(T)^\dagger W|j'\rangle,$$

where P_n is the projection onto the subspace where the unitary transformation W is considered.

The theory of quantum gates uses the *Cartan decomposition* on the SU(4) group, the *Weyl chamber*, and *local invariants* related to the *equivalence classes* [79], [88], [93] (Calarco, Koch, Müller, Reich, Vala, and others). The Cartan decomposition for a two-qubit operator $U \in \mathrm{SU}(4)$ is

$$U = k_1 A k_2, \quad \text{where } A = \exp\left(\frac{i}{2}(c_1\sigma_x\otimes\sigma_x + c_2\sigma_y\otimes\sigma_y + c_3\sigma_z\otimes\sigma_z)\right) \tag{2.21}$$

(see [88]). Here σ_x, σ_y, σ_z are the Pauli matrices, $k_1, k_2 \in \mathrm{SU}(2)\otimes\mathrm{SU}(2)$ are local operations, and the real numbers c_1, c_2, c_3 are coordinates in the Weyl chamber. W_{CNOT} can be represented by (2.21) as

$$W_{\mathrm{CNOT}} = (\mathbb{I}\otimes W_{\mathrm{H}})W_{\mathrm{CPHASE}}(\mathbb{I}\otimes W_{\mathrm{H}}),$$

where $\mathbb{I}$ is the identity matrix [79]. W_{CNOT} and W_{CPHASE} differ from each other only by local operations. Hence, they are locally equivalent ($W_{\mathrm{CNOT}} \sim W_{\mathrm{CPHASE}}$) and belong to the same equivalence class $[W_{\mathrm{CNOT}}]$, which corresponds to the point $(c_1, c_2, c_3) = (\pi/2, 0, 0)$ in the Weyl chamber. The equivalence classes are connected with local invariants that can be written using the following representation of U via the Bell basis [88]:

$$g_1 = \frac{1}{16}\operatorname{Re}\{(\operatorname{Tr}\widehat{m})^2\}, \qquad g_2 = \frac{1}{16}\operatorname{Im}\{(\operatorname{Tr}\widehat{m})^2\}, \qquad g_3 = \frac{1}{4}[(\operatorname{Tr}\widehat{m})^2 - \operatorname{Tr}\widehat{m}^2],$$

where

$$\widehat{m} = U_B^\dagger U_B, \qquad U_B = BUB^\dagger, \qquad B = \frac{1}{2}\begin{pmatrix} 1 & 0 & 0 & i \\ 0 & i & 1 & 0 \\ 0 & i & -1 & 0 \\ 1 & 0 & 0 & -i \end{pmatrix}.$$

The papers [79], [88], and [93] consider OCPs where the cost functional is formulated in terms of distances in the space of coordinates (c_1, c_2, c_3) or in the space of local invariants (g_1, g_2, g_3) for a given target gate W_{target}. The terminant [88]

$$\begin{aligned} \mathscr{F}_{\mathrm{LI}} &= \sum_{i=1}^{3}(\Delta g_i)^2 + 1 - \frac{1}{n}\operatorname{Tr}\{U_n(T)U_n^\dagger(T)\}, \\ \Delta g_i &= |g_i(W_{\mathrm{target}}) - g_i(U_n(T))|, \end{aligned} \tag{2.22}$$

is considered, where $U_n(T) = P_n U(T) P_n$ is the result of projecting onto a subspace. The explicit form of (2.22) in terms of $\{\psi_j(T)\}_{j=1}^n$ is a polynomial of degree eight. The corresponding optimisation results [89] are described in Section 5.

2.4. Gross–Pitaevskii equation and optimal control of Bose–Einstein condensates. The linear Schrödinger equation is often suitable for describing quantum systems, but in some situations non-linear equations appear, such as the Gross–Pitaevskii equation proposed by Gross and Pitaevskii in 1961. This equation describes the dynamics of a Bose–Einstein condensate, which is important both from the theoretical point of view and for creation of new technologies, for example, atomic chips [121]. Modelling of the controlled dynamics of a Bose–Einstein condensate is performed using the Gross–Pitaevskii equation with control ([23], [40], [66], [97], [98], [121]–[126]).

Definition 2.10. The equation

$$\frac{\partial \psi(x,t)}{\partial t} = -\frac{i}{\hbar}\big(K + V(x,u(t)) + \kappa|\psi(x,t)|^2\big)\psi(x,t) \tag{2.23}$$

with initial condition $\psi(x,0) = \psi_0(x)$ is called the Gross–Pitaevskii equation with control $u \in \mathscr{U}$ in the potential (see, for example, [23], p. 336). Here $x \in \Omega \subseteq \mathbb{R}^d$, $\psi(\,\cdot\,,t) \in L^2(\Omega;\mathbb{C})$, $K = -\dfrac{\hbar^2}{2m}\nabla^2$ is the kinetic-energy operator, m is the atomic mass, $V(x,u(t))$ is the controlled potential, κ is a coefficient (for example, $\kappa = U_0(N_a - 1)$ [40] where N_a is the number of atoms and U_0 is the strength of interaction between atoms for one-dimensional x), and the class $\mathscr{U}$ is defined in (2.3).

For brevity we will write $\psi(t)$ to mean $\psi(\,\cdot\,,t)$.

Authors use potentials of various forms ([23], [40], [66], [97], [98], [121]–[126]). In [66] (Sklarz and Tannor, 2002) the Gross–Pitaevskii equation (2.23) was considered with the following potential for one-dimensional x:

$$V(t,x,u(t)) = u(t)x^2 + s(t)V_0\cos^2(kx). \tag{2.24}$$

Here the control u characterises the strength of the trap potential, V_0 characterises the lattice intensity, $s(t)$ is the function governing the switching-on of the field, and k is the laser field wavenumber. The wavepackets represent quantum bits for quantum information. Initially, the Bose–Einstein condensate is located in the ground state of the potential. The OCP is considered with the condition

$$J(v) = \langle\psi(T), (\cos\theta(T))^2\psi(T)\rangle - \langle\psi(T), \cos\theta(T)\psi(T)\rangle^2 \to \min, \tag{2.25}$$

where $\theta(T) = \theta(\,\cdot\,,T)$ is the phase of the wavepacket at time T, $\cos\theta = \dfrac{1}{2}\dfrac{\psi+\psi^*}{|\psi|} = \dfrac{\operatorname{Re}\psi}{|\psi|}$, and $v = (\psi,u)$.

In [122] the following potentials for one-dimensional x were considered:

$$V(x,u(t)) = \frac{1}{2}\big(x - u(t)x_0\big)^2, \tag{2.26}$$

and

$$V(x,u(t)) = \begin{cases} \dfrac{1}{2}\left(|x| - u(t)\dfrac{d}{2}\right)^2, & |x| > u(t)\dfrac{d}{4}, \\ \dfrac{1}{2}\left(\dfrac{(u(t))^2}{8} - x^2\right), & |x| \leqslant u(t)\dfrac{d}{4}. \end{cases} \tag{2.27}$$

In [98], [121], and [123] the following polynomial potential for one-dimensional x was used:

$$V(t,x,u(t)) = p_2\big(x-u(t)\big)^2 + p_4\big(x-u(t)\big)^4 + p_6\big(x-u(t)\big)^6. \tag{2.28}$$

Here the control u defines the motion along the axis Ox to shake the condensate, and the numbers p_2, p_4, and p_6 are fitting parameters. The condensate is initially prepared in the ground state $V(x,0,u(0))$ with interacting bosons, and ψ_{target} is set to be the first excited state for V.

The common goal of control is to maximise the probability $|\langle\psi_{\text{target}},\psi(T)\rangle_{L^2}|^2$ of a transition to the target state ψ_{target}. This control goal is described by the terminants (2.5) and (2.6) for (2.23). Sometimes, together with the GRAPE method ([39], [40]), which will be described in Section 6, the following H_1-regulariser is included in the cost functional for smoothing of the control:

$$\lambda_{du}\int_0^T (\dot u(t))^2\,dt, \qquad \lambda_{du} > 0. \tag{2.29}$$

For (2.23), consider the general cost functional to be minimised:

$$J(v) = \mathscr{F}(\psi(T)) + \lambda_u\int_0^T \frac{\|u(t)\|^2}{S(t)}\,dt + \lambda_{du}\int_0^T (\dot u(t))^2\,dt \to \min, \tag{2.30}$$

where $\mathscr{F}$ is defined, for example, using (2.5), $\lambda_u, \lambda_{du} \geqslant 0$ are parameters, and S is a shape function.

A rigorous formulation of the optimal control problem for the Gross–Pitaevskii equation with the potential $V(x,u) = U(x) + u(t)\widetilde V(x)$, where U and $\widetilde V$ are given and $u(t)$ is the control, is provided in [124]. In this formulation the potential V is in the space $W^{1,\infty}(\mathbb{R}^d)$ of Lipschitz functions, and the potential U satisfies the conditions

$$U \in C^\infty(\mathbb{R}^d)$$

and

$$\partial^k U \in L^\infty(\mathbb{R}^d)$$

for any multi-index k such that $|k| > 2$. The initial state $\psi_0(x)$ belongs to the subspace

$$\Upsilon = \{\psi \in H^1(\mathbb{R}^d)\colon x\psi \in L^2(\mathbb{R}^d)\}.$$

The cost functional has the form

$$J(\psi,u) = \langle\psi(T,\,\cdot\,), A\psi(T,\,\cdot\,)\rangle_{L^2(\mathbb{R}^d)} + \int_0^T (\dot u(t))^2\left[\gamma_1\left(\int_{\mathbb{R}^d}\widetilde V(x)|\psi(x,t)|^2\,dx\right)^2 + \gamma_2\right]dt,$$

where $T > 0$, $\gamma_1 \geqslant 0$, $\gamma_2 > 0$, and $A\colon \Upsilon \to L^2(\mathbb{R}^d)$ is an operator acting in $L^2(\mathbb{R}^d)$ which is essentially self-adjoint (and possibly unbounded). The integral terms in the cost functional are introduced to avoid highly oscillating control. For this control problem, the well-posedness and existence of an optimal control were proved.

2.5. Controllability of closed quantum systems. The problem of constructing optimal quantum controls is closely related to the ideas behind and criteria for controllability of quantum systems as discussed, for example, in the articles [127]–[134] and the monographs [11] and [23]. Before starting to search for an optimal control, it is desirable to know whether such a control exists at all. The answer to this question is given by the controllability criteria, which are well known for closed quantum systems. Below we will give the basic concepts and results without details.

For closed systems one fundamental notion is the concept of projective state controllability or equivalent state controllability.

Definition 2.11. A quantum system (2.1)–(2.3) with states $\psi(t) \in \mathbb{C}^n$ is said to be projective state controllable if for any initial state ψ_0 and final equivalence class

$$[\psi_{\text{target}}] := \{e^{i\phi}\psi_{\text{target}}\colon \phi \in [0, 2\pi)\}$$

with some ψ_{target} there exist a $T > 0$ and a control $u \in \mathscr{U}$ such that the system can be moved from ψ_0 to $[\psi_{\text{target}}]$ in time T.

Also important is the notion of controllability on the special unitary group.

Definition 2.12. A system (2.12) describing the evolution of a unitary operator $U(t) \in \mathbb{C}^{n\times n}$ is said to be controllable on the group $\mathrm{SU}(n)$ if for any unitary operator $W \in \mathrm{SU}(n)$ there exist a time $T > 0$ and a control $u \in \mathscr{U}$ such that $W = e^{i\phi}U(T)$, where $\phi \in [0, 2\pi)$ is some phase.

The analysis of controllability of quantum systems, including the systems (2.1)–(2.3), is crucial for quantum control, since the presence or absence of controllability determines the solvability of an OCP. For example, the terminant $\mathscr{F}_{\psi_{\text{target}}}$ (2.5) determines the probability for a system state transfer on the sphere over the time T. Realisation of this transfer between arbitrary initial and target states is impossible for uncontrollable systems.

The control criteria are based on the analysis of the Lie algebra $\mathscr{L}$ generated by all possible commutators of the operators $\mathbf{H}_0, \mathbf{H}_1, \ldots, \mathbf{H}_m$. Without loss of generality we can set $\operatorname{Tr}\mathbf{H}_i = 0$ for $i = 0, \ldots, m$. Indeed, the dynamics of a system with operators having non-zero traces will differ from the dynamics with the operators $\mathbf{H}_i - \operatorname{Tr}\mathbf{H}_i/n$ by a physically unessential phase factor. One consequence of the analysis in [118] is the following theorem [135], [136].

Theorem 2.2. *Let $\mathscr{H} = \mathbb{C}^n$. The system (2.12) is projective state controllable if and only if the Lie algebra $\mathrm{Lie}\{-i\mathbf{H}_0, \ldots, -i\mathbf{H}_m\}$ generated by all commutators of the operators $-i\mathbf{H}_0, \ldots, -i\mathbf{H}_m$ is isomorphic to the Lie algebra $\mathfrak{sp}(n/2)$ or $\mathfrak{su}(n)$ when n is even, and to the Lie algebra $\mathfrak{su}(n)$ when n is odd.*

Verification of the controllability for a particular quantum system is one of the basic problems coming before the search for optimal controls. Projective state controllability of the system (2.1)–(2.3) means that there exists a time T and a control

$u \in \mathscr{U}$ such that $\mathscr{F}_{\psi_{\text{target}}} = 0$ (exact controllability). For some sets Q and some values of T, the system may not be controllable. On the other hand, we can look for Q and T such that $\mathscr{F}_{\psi_{\text{target}}}(\psi(T)) = 0$ and some additional condition is satisfied, for example, one can require that T takes the minimum possible value.

2.6. Landscapes of control problems for closed quantum systems. The goal of applying numerical methods to quantum control problems is to find globally optimal controls, those that, for a minimisation problem, deliver a global minimum to the cost functional. If all the minima of the cost functional were global, then the most natural numerical methods for finding optimal controls would be methods of gradient type. If the system is controllable, but the landscape is replete with traps (that is, local but not global minima), then stochastic algorithms (for example, genetic algorithms) would be an appropriate choice in order to step around or out of such traps. Therefore, a theoretical analysis of minima of the cost functionals is important when choosing an appropriate numerical strategy for finding optimal controls. Here we briefly outline some results on this topic.

The problem of analysing extrema of cost functionals for quantum systems was posed in [137] (Rabitz, Hsieh, and Rosenthal, 2004), where the conjecture was made that all the minima of cost functionals are global (that is, control landscapes are free of traps). Since then, gradient-based methods have frequently been found to give good results in quantum-control numerical simulations and experiments. A large number of simulations have shown that even though the landscapes for optimal control problems may contain singular critical points and traps, the conditions necessary to produce such points are sufficiently stringent that many control landscapes may lack traps [138], [139].

A considerable effort has been made to understand the effectiveness of gradient-based optimal control simulations. However, despite this, the trap-free assumption has been rigorously proven only for quantum systems with $n = 2$ levels (Theorem 2.3) [119], [120] (Pechen and Il'in, 2012, 2014) and for the problem of controlling the passage of a quantum particle through a one-dimensional potential barrier (Theorem 2.4) [140] (Pechen and Tannor, 2014). For dimension greater than two, systems with trapping properties have been found [141] (Pechen and Tannor, 2011), [142] (de Fouquieres and Schirmer, 2013). Numerical evidence for trap-free behaviour in various cases was emphasised in [143] (Rabitz, Ho, Long, Wu, and Brif, 2012).

Let $\lambda_u = 0$ in (2.14). Then the cost functional is $J_X(u) = \mathscr{F}_X(U(T))$, where the terminant $\mathscr{F}_X \colon \mathrm{SU}(n) \to \mathbb{R}$ is a function on the special unitary group and $U(T)$ is the solution of the Schrödinger equation at time T for the control u. For example, for (2.15) one has

$$\mathscr{F}_W(U) = -\frac{1}{n^2} |\operatorname{Tr}(W^\dagger U)|^2.$$

Definition 2.13. The graph of the functional $J_X(u)$ is called the control landscape of the control problem. The control u is called a trap if it gives a local but not a global minimum for $J_X(u)$.

For single-qubit gates ([119], [120], [144], [145]) let us consider control landscapes for systems of the form (2.12) with $U(t) \in \mathbb{C}^{2\times 2}$, scalar control ($m = 1$), and cost

functionals J_O and J_W, which describe the mean value of an observable and the realisation of a single-qubit gate. The main result is the following theorem [120].

Theorem 2.3. *For the system (2.12) with $\mathscr{H} = \mathbb{C}^2$ and a scalar control ($m = 1$) let the operators $\mathbf{H}_0$, $\mathbf{H}_1$ and the time T be such that $[\mathbf{H}_0, \mathbf{H}_1] \neq 0$ and*

$$T \geqslant \frac{\pi}{\|\mathbf{H}_0 - \operatorname{Tr}\mathbf{H}_0/2 + u_0(\mathbf{H}_1 - \operatorname{Tr}\mathbf{H}_1/2)\|},$$

where

$$u_0 := \frac{-\operatorname{Tr}(\mathbf{H}_0)\operatorname{Tr}(\mathbf{H}_1) + 2\operatorname{Tr}(\mathbf{H}_0\mathbf{H}_1)}{(\operatorname{Tr}\mathbf{H}_1)^2 - 2\operatorname{Tr}\mathbf{H}_1^2}.$$

Then all the minima of the cost functionals

$$J_O(u) = \langle O\rangle_T = \operatorname{Tr}\{U(T)\rho_0 U^\dagger(T)O\} \quad \text{and} \quad J_W(u) = -\frac{1}{4}|\operatorname{Tr}(W^\dagger U(T))|^2$$

are global minima for any Hermitian observable O, any unitary operator W, and any density matrix ρ_0.

The problem of controlling the tunneling of a quantum particle through a potential barrier is described by the stationary Schrödinger equation

$$\left(-\frac{d^2}{dx^2} + V(x)\right)\Psi(x) = E\Psi(x), \qquad E \in \mathbb{R}.$$

The potential $V(x)$ is assumed to have compact support ($V(x) = 0$ if $|x| > a$ for some $a > 0$). The solution that has the following asymptotics is considered:

$$\Psi(x) = \begin{cases} e^{i\sqrt{E}\,x} + A_E e^{-i\sqrt{E}\,x}, & x < -a, \\ B_E e^{i\sqrt{E}\,x}, & x > a. \end{cases}$$

This solution describes a particle falling from the left on the potential, being reflected from the potential barrier with probability $|A_E|^2$, and passing to the right through the barrier with probability $T_E(V) = |B_E|^2$. The potential is regarded as the control. The transmission coefficient $T_E(V)$ is the cost functional that is to be maximised. The main result is the following theorem [140].

Theorem 2.4. *All the extrema of the transmission coefficient $T_E(V) = |B_E|^2$ are global maxima corresponding to the value $T_E(V) = 1$ (complete tunneling).*

Thus, the absence of traps for quantum systems is proved for two-dimensional and infinite-dimensional Hilbert spaces. In the general case of an arbitrary dimension, the problem remains open. As a result, it is important to develop effective methods for obtaining optimal controls for quantum systems of any dimension.

3. Krotov method for systems with states in $\mathbb{R}^n$

3.1. Optimal control problems and the Krotov Lagrangian. Consider the following class of OCPs with real-valued states.

Definition 3.1. The following problem is called an OCP with free final state for a dynamical system defined by an ODE with a control:

$$\frac{dy(t)}{dt} = f(t, y(t), u(t)), \qquad y(0) = y_0, \tag{3.1}$$

$$u \in \mathscr{U} = \mathrm{PC}([0, T]; Q), \qquad Q \subseteq \mathbb{R}^m, \tag{3.2}$$

$$J(v) = \mathscr{F}(y(T)) + \int_0^T f^0(t, y(t), u(t))\, dt \to \inf_{v \in \mathscr{D}}, \qquad v = (y, u), \tag{3.3}$$

where y is a continuous, piecewise differentiable function and $\mathscr{D}$ is the set of admissible processes $v = (y, u)$. The state y_0 and the time T are fixed. For $\mathscr{F}$, f^0, and f the following conditions are traditional:

1) the vector function $f(t, y, u) = (f_1(t, y, u), \ldots, f_n(t, y, u))$ and the scalar function $f^0(t, y, u)$, together with their partial derivatives with respect to y and u, are defined and continuous on the set of variables $(t, y, u) \in [0, T] \times \mathbb{R}^n \times Q$, and the vector function f satisfies a Lipschitz condition with respect to y, that is,

$$\|f(t, y + \Delta y, u)\| \leqslant L\|\Delta y\| \qquad \forall\, u \in Q, \quad t \in [0, T];$$

2) the function $\mathscr{F}(y)$ is continuously differentiable on $\mathbb{R}^n$.

As is known (see [146], for example), for any control $u \in \mathscr{U}$ the Cauchy problem (3.1) has a unique solution y in the class of piecewise differentiable functions. Continuous differentiability of f^0 and f with respect to u is also necessary for further considerations.

Definition 3.2. A solution of the problem (3.1)–(3.3) is understood to be a *minimising sequence*, that is, a sequence of processes $\{v^{(k)}\}_{k \geqslant 0}$ in $\mathscr{D}$ that satisfies the relation

$$\lim_{k \to \infty} J(v^{(k)}) = \inf_{v \in \mathscr{D}} J(v).$$

If there is an element $\overline{v} \in \mathscr{D}$ such that $J(\overline{v}) = \min_{v \in \mathscr{D}} J(v)$, then the process $\overline{v}$ and the control $\overline{u}$ are said to be (globally) *optimal*.

Definition 3.3. For the problem (3.1)–(3.3) consider a process $v^{(k)} \in \mathscr{D}$ ($k \geqslant 0$ is the iteration number). The determination of a process $v^{(k+1)} \in \mathscr{D}$ for which $J(v^{(k+1)}) < J(v^{(k)})$ is called an improvement of $v^{(k)}$. A sequence $\{v^{(k)}\}_{k=0}^{K} \subset \mathscr{D}$ such that $J(v^{(k+1)}) < J(v^{(k)})$ for $k = 0, \ldots, K$ is called an improving sequence.

In the early 1960s Krotov introduced the concept of the *generalised Lagrangian* (later called the *Krotov Lagrangian*) and formulated *sufficient conditions for optimality* [59], [60].

Definition 3.4. For the problem (3.1)–(3.3), the Krotov Lagrangian is the functional

$$L^{\varphi}(v) = G^{\varphi}(y(T)) - \int_0^T R^{\varphi}(t, y(t), u(t))\, dt, \qquad v \in \mathscr{E}, \tag{3.4}$$

where $\mathscr{E}$ is an extension of the set $\mathscr{D}$,

$$G^{\varphi}(y(T)) = \mathscr{F}(y(T)) + \varphi(T, y(T)) - \varphi(0, y(0)), \tag{3.5}$$

$$R^{\varphi}(t, y, u) = \left\langle \frac{\partial \varphi(t, y)}{\partial y}, f(t, y, u) \right\rangle - f^0(t, y, u) + \frac{\partial \varphi(t, y)}{\partial t}. \tag{3.6}$$

Here φ belongs to the set Φ of functions each of which has continuous partial derivatives for all t and y except possibly at a finite number of values of t on $[0,T]$.

As is known ([59], [60]), $L^{\varphi}(v) \equiv J(v)$ on $\mathscr{D}$ for any $\varphi \in \Phi$, and thus $L(v)$ can be considered as a special representation of $J(v)$. The partial derivatives of $\varphi(t,y)$ with respect to y are Lagrange multipliers. The function R defined in (3.6) can be written as

$$R^{\varphi}(t,y,u) = H\left(t, \frac{\partial \varphi(t,y)}{\partial y}, y, u\right) + \frac{\partial \varphi(t,y)}{\partial t},$$

where

$$H(t,q,y,u) = \langle q, f(t,y,u)\rangle - f^0(t,y,u)$$

is the Pontryagin function, $y \in \mathbb{R}^n$, and $u \in Q \subseteq \mathbb{R}^m$.

The following problem is called an OCP for a bilinear system with regularisation with respect to the control (see [147], for instance):

$$\frac{dy(t)}{dt} = \left(A(t) + \sum_{j=1}^{m} u_j(t) B_j(t)\right) y(t), \qquad y(0) = y_0, \tag{3.7}$$

$$J(v) = \mathscr{F}(y(T)) + \lambda_u \int_0^T \sum_{j=1}^{m} u_j^2(t)\, dt \to \inf_{v \in \mathscr{D}}, \qquad \lambda_u \geqslant 0, \tag{3.8}$$

where the control u belongs to the class $\mathscr{U}$ defined in (3.2).

The problem (2.1)–(2.4) with states $\psi(t) \in \mathbb{C}^n$ can be described by the corresponding OCP with states $y(t) \in \mathbb{R}^{2n}$ if the vector $y(t)$ represents both the real and the imaginary parts of a complex-valued $\psi(t)$:

$$y_j(t) = \operatorname{Re} \psi_j(t) \quad \text{and} \quad y_{n+j}(t) = \operatorname{Im} \psi_j(t) \quad \text{for } j = 1, \ldots, n.$$

For example, the problem (3.7), (3.8) with $\mathscr{F}(y(T)) = -\langle y(T), My(T)\rangle$, $M \geqslant 0$, can describe certain quantum OCPs. The terminant $\mathscr{F}(y(T)) = -\langle y(T), My(T)\rangle$ is bounded below due to the invariant

$$\|y(t)\|^2_{\mathbb{R}^{2n}} = \|\psi(t)\|^2_{\mathbb{C}^n} = 1.$$

3.2. Krotov method in the general form. For the problem (3.1)–(3.3), consider the iterative process with $v^{(k)} = (y^{(k)}, u^{(k)})$ and $v^{(k+1)} = (y^{(k+1)}, u^{(k+1)})$ the input and output admissible processes, respectively, at the kth iteration of the method.

1. Compute the function $\varphi^{(k)} \in \Phi$ which satisfies the conditions[6]

$$G^{\varphi^{(k)}}(y^{(k)}(T)) = \max_{y \in \mathbb{R}^n} G^{\varphi^{(k)}}(y), \tag{3.9}$$

$$R^{\varphi^{(k)}}(t, y^{(k)}(t), u^{(k)}(t)) = \min_{y \in \mathbb{R}^n} R^{\varphi^{(k)}}(t, y, u^{(k)}(t)), \qquad t \in [0, T). \tag{3.10}$$

2. Find the solution $y^{(k+1)}$ of the Cauchy problem

$$\frac{dy^{(k+1)}(t)}{dt} = f\big(t, y^{(k+1)}(t), \widetilde{u}^{(k)}(t, y^{(k+1)}(t))\big), \qquad y^{(k+1)}(0) = y_0, \tag{3.11}$$

and find the control $u^{(k+1)}$ defined by

$$u^{(k+1)}(t) = \widetilde{u}^{(k)}(t, y^{(k+1)}(t)) := \arg\max_{u \in Q} R^{\varphi^{(k)}}\big(t, y^{(k+1)}(t), u\big), \qquad t \in [0, T]. \tag{3.12}$$

Theorem 3.1. *For the problem* (3.1)–(3.3), *the method* (3.9)–(3.12) *provides a sequence of processes* $\{v^{(k)}\} \subset \mathscr{D}$ *such that* $J(v^{(k+1)}) \leqslant J(v^{(k)})$. *Moreover, if*

$$\int_0^T \max_{u \in Q} R^{\varphi^{(k)}}(t, y^{(k)}(t), u)\, dt \neq \int_0^T R^{\varphi^{(k)}}(t, y^{(k)}(t), u^{(k)}(t))\, dt, \tag{3.13}$$

then $\{v^{(k)}\}$ *is an improving sequence.*

Proof. Following [58], to prove the theorem consider the increment $J(v^{(k)}) - J(v)$ for a given process $v^{(k)} \in \mathscr{D}$, an arbitrary process $v \in \mathscr{D}$, and $\varphi \in \Phi$:

$$J(v^{(k)}) - J(v) = L^{\varphi}(v^{(k)}) - L^{\varphi}(v) = G^{\varphi}(y^{(k)}(T)) - G^{\varphi}(y(T)) + \int_0^T \big(R^{\varphi}(t, y(t), u(t)) - R^{\varphi}(t, y^{(k)}(t), u^{(k)}(t))\big)\, dt. \tag{3.14}$$

To satisfy the inequality $J(v^{(k)}) \geqslant J(v)$ for $v \in \mathscr{D}$ it is enough to find a function $\varphi = \varphi^{(k)} \in \Phi$ such that its values $\varphi^{(k)}(t, y^{(k)}(t))$ and $\varphi^{(k)}(t, y(t))$ on $[0, T]$ satisfy

$$G^{\varphi^{(k)}}(y^{(k)}(T)) - G^{\varphi^{(k)}}(y(T)) \geqslant 0, \tag{3.15}$$

$$R^{\varphi^{(k)}}(t, y(t), u(t)) - R^{\varphi^{(k)}}(t, y^{(k)}(t), u^{(k)}(t)) \geqslant 0, \tag{3.16}$$

[6]Here

$$R^{\varphi^{(k)}}(t, y^{(k)}(t), u^{(k)}(t)) = \left\langle \frac{\partial \varphi^{(k)}(t, y^{(k)}(t))}{\partial y}, f(t, y^{(k)}(t), u^{(k)}(t)) \right\rangle - f^0(t, y^{(k)}(t), u^{(k)}(t)) + \frac{\partial \varphi^{(k)}(t, y^{(k)}(t))}{\partial t},$$

where $\dfrac{\partial \varphi^{(k)}(t, y^{(k)}(t))}{\partial y} = \dfrac{\partial \varphi^{(k)}(t, y)}{\partial y}\Big|_{y = y^{(k)}(t)}$ and $\dfrac{\partial \varphi^{(k)}(t, y^{(k)}(t))}{\partial t} = \dfrac{\partial \varphi^{(k)}(t, y)}{\partial t}\Big|_{y = y^{(k)}(t)}$. In this survey, for brevity we use notation like $\dfrac{\partial \varphi^{(k)}(t, y^{(k)}(t))}{\partial y}$ instead of $\dfrac{\partial \varphi^{(k)}(t, y)}{\partial y}\Big|_{y = y^{(k)}(t)}$.

and for $J(v^{(k)}) > J(v)$ it is sufficient to have strict inequality in (3.16) on some subset of $[0,T]$ of positive measure. Splitting the left-hand side of (3.16) into the sum of two partial increments, we obtain the following conditions:

$$R^{\varphi^{(k)}}(t,y(t),u(t)) - R^{\varphi^{(k)}}(t,y(t),u^{(k)}(t)) \geqslant 0, \tag{3.17}$$

$$R^{\varphi^{(k)}}(t,y(t),u^{(k)}(t)) - R^{\varphi^{(k)}}(t,y^{(k)}(t),u^{(k)}(t)) \geqslant 0. \tag{3.18}$$

To compute $\varphi^{(k)}$, consider the conditions (3.9) and (3.10) obtained from (3.15) and (3.18). Because of (3.17), for each t we consider the condition

$$R^{\varphi^{(k)}}(t,y(t),u(t)) = \max_{u\in Q} R^{\varphi^{(k)}}(t,y(t),u),$$

which leads to the formula (3.12), where the process $v=(y,u)$ satisfying the above conditions is denoted by

$$v^{(k+1)} = (y^{(k+1)}, u^{(k+1)}).$$

Thus, to find the function $y^{(k+1)}$ we integrate the system (3.11) derived from (3.1), where $u(t) = \widetilde{u}^{(k)}(t, y^{(k+1)}(t))$. Because of possible discontinuities of $\widetilde{u}^{(k)}(t,y)$ with respect to y, integration of (3.11) is in general carried out using the theory of ODEs with discontinuous right-hand side [148]. We suppose that there is a triple $(\varphi^{(k)}, y^{(k+1)}, u^{(k+1)})$ satisfying (3.10)–(3.11). Then the process $v^{(k+1)}$ gives either $J(v^{(k+1)}) < J(v^{(k)})$ (an improvement) or $J(v^{(k+1)}) = J(v^{(k)})$. If (3.13) is not satisfied, then $J(v^{(k+1)}) = J(v^{(k)})$. □

Solving the sequence of improvement problems via (3.9)–(3.12), we obtain a control sequence:

$$u^{(0)} \to u^{(1)} \to \cdots \to u^{(k)} \to \cdots \to u^{(K)}.$$

Consider the problem (3.7), (3.8). The Pontryagin function is

$$H(t,q,y,u) = \left\langle q, A(t) + \sum_{j=1}^{m} u_j B_j(t) \right\rangle - \lambda_u \sum_{j=1}^{m} u_j^2, \qquad q = \frac{\partial \varphi}{\partial y}(t,y).$$

Let $m=1$, $Q=[a,b]$, and $\lambda_u = 0$. In this case, the Pontryagin function is linear in u and the function $\widetilde{u}^{(k)}$ defined in (3.12) is

$$\widetilde{u}^{(k)}(t,y) = \begin{cases} a, & g^{(k)}(t,y) < 0, \\ u_{\text{sing}}^{(k)}(t,y) \in [a,b], & g^{(k)}(t,y) = 0, \\ b, & g^{(k)}(t,y) > 0, \end{cases} \tag{3.19}$$

where

$$g^{(k)}(t,y) = \left\langle \frac{\partial \varphi^{(k)}(t,y)}{\partial y}, B(t)y \right\rangle$$

is called the switching function, and the subscript 'sing' means 'singular'. Substituting (3.19) into (3.7), we obtain a system of ODEs which is in general discontinuous with respect to y. Therefore, the conditions for the existence and uniqueness of the solution of the Cauchy problem are violated [148].

Further, consider $m = 1$, $Q = [a, b]$, and $\lambda_u > 0$. The function $\widetilde{u}^{(k)}$ has the following form:

$$\widetilde{u}^{(k)}(t,y) = \begin{cases} a, & u_{\mathrm{st}}^{(k)}(t,y) < a, \\ u_{\mathrm{st}}^{(k)}(t,y), & u_{\mathrm{st}}^{(k)}(t,y) \in [a,b], \\ b, & u_{\mathrm{st}}^{(k)}(t,y) > b, \end{cases} \tag{3.20}$$

where $u_{\mathrm{st}}^{(k)}(t,y)$ is a stationary point obtained from the condition $\partial H/\partial u = 0$ for the Pontryagin function, and the subscript 'st' means 'stationary point'. The function (3.20) is continuous with respect to y.

3.3. Krotov method with linear-quadratic function φ. Based on the articles [58] and [61], we describe the second-order Krotov method ($\varphi(t,y)$ is taken in the class of linear-quadratic functions) for the problem (3.1)–(3.3).

Definition 3.5. The function $\varphi(t,y)$ is said to be linear-quadratic if it has the form

$$\varphi(t,y) = \langle p(t), y\rangle + \frac{1}{2}\big\langle y - y^{(k)}(t), \Sigma(t)(y - y^{(k)}(t))\big\rangle \tag{3.21}$$

with some continuous, piecewise differentiable vector function $p = (p_1, \dots, p_n)$ and the matrix function $\Sigma = (\Sigma_{i,j})_{i,j=1,\dots,n}$.

According to [61] we have the following result.

Lemma 3.1. *Consider the problem* (3.1)–(3.3). *Suppose that*:
1) *for the functions* $\mathscr{F}(y)$ *and* $f^0(t,y,u)$ *there exist constants* $K, M < \infty$ *such that for all* $y \in R^n$ *with* $\|y\| \geqslant M$

$$\mathscr{F}(y) \leqslant K\|y\|^2, \qquad f^0(t,y,u) \leqslant K\|y\|^2 \quad \forall\, (t,u) \in [0,T] \times Q;$$

2) *for the function* $f(t,y,u)$ *there exist* $K, M < \infty$ *such that for all* $(t,y,u) \in [0,T] \times \mathbb{R}^n \times Q$ *with* $\|y\| \geqslant M$

$$f(t,y,u) \leqslant K\|y\|;$$

3) *for the Jacobi matrix of* $f = (f_1, \dots, f_n)$ *there exists an* $A < \infty$ *such that for all* $(t,y,u) \in [0,T] \times R^n \times Q$

$$\left\| \frac{\partial f_i(t,y,u)}{\partial y_j} \right\| \leqslant A,$$

where $\|\cdot\|$ *is the matrix norm.*

Then for the process $v^{(k)} = (y^{(k)}, u^{(k)}) \in \mathscr{D}$ *there is a solution* $\varphi^{(k)}(t,y)$ *of the problem* (3.9), (3.10) *in the form* (3.21), *where the function* $p = p^{(k)}$ *is defined as the solution of the Cauchy problem*

$$\begin{gathered} \frac{dp^{(k)}(t)}{dt} = -\frac{\partial H}{\partial y}(t, p^{(k)}(t), y^{(k)}(t), u^{(k)}(t)), \\ p^{(k)}(T) = -\frac{\partial \mathscr{F}}{\partial y}(y^{(k)}(T)) \end{gathered} \tag{3.22}$$

(*the system is integrated 'from right to left'*), *and the matrix function* $\Sigma = \Sigma^{(k)}$ *is defined by*

$$\Sigma^{(k)}(t) = \big(\alpha(e^{\gamma(T-t)} - 1) + \beta\big)\mathbb{I}_n, \tag{3.23}$$

where the values $\alpha, \beta < 0$ *and* $\gamma > 0$ *are given and* $\mathbb{I}_n$ *is the identity matrix.*

For the problem (3.1)–(3.3), consider the following iterative process, where $v^{(k)} = (y^{(k)}, u^{(k)})$ and $v^{(k+1)} = (y^{(k+1)}, u^{(k+1)})$ are the input and output admissible processes, respectively, at the kth iteration of the method.

1. Consider a linear-quadratic function $\varphi^{(k)}$ with $\Sigma^{(k)}$ defined by (3.23) for some values of $\alpha, \beta < 0$ and $\gamma > 0$, and find $p^{(k)}$ as the solution of the Cauchy problem (3.22).
2. Find the solution $y^{(k+1)}$ of the Cauchy problem (3.11). Find the control $u^{(k+1)}$ defined by

$$\begin{aligned} u^{(k+1)}(t) &= \widetilde{u}^{(k)}(t, y^{(k+1)}(t)) \\ &:= \arg\max_{u \in Q} H\big(t, p^{(k)}(t) + \Sigma^{(k)}(t)(y^{(k+1)}(t) - y^{(k)}(t)), y^{(k+1)}(t), u\big). \end{aligned} \tag{3.24}$$

Theorem 3.2. *For the problem* (3.1)–(3.3) *let the conditions* 1)–3) *in Lemma* 3.1 *be satisfied. Then the method* (3.11), (3.21)–(3.24) *provides a sequence of processes* $\{v^{(k)}\} \subset \mathscr{D}$ *such that* $J(v^{(k+1)}) \leqslant J(v^{(k)})$. *Moreover, if*

$$\int_0^T \max_{u \in Q} H(t, p^{(k)}(t), y^{(k)}(t), u)\, dt \neq \int_0^T H(t, p^{(k)}(t), y^{(k)}(t), u^{(k)}(t))\, dt, \tag{3.25}$$

then $\{v^{(k)}\}$ *is an improving sequence.*

If for some selected values $\alpha, \beta < 0$ and $\gamma > 0$ there is no improvement, then one has to adjust these parameters by decreasing α and β and increasing γ (it is also possible to change only one of these parameters), and then, without changing the iteration index, calculate the output process $v^{(k+1)}$ for the updated values of these parameters [61]. If several re-selections of parameters do not give an improvement, then we stop the iterations.

In (3.9), (3.10) we take the global minimum and maximum. Nevertheless, in order to simplify the general Krotov method, it was proposed to use the first- and second-order necessary conditions for *local* extrema in (3.9), (3.10) [58]. Consider the equations

$$R_y^{\varphi^{(k)}}(t, y^{(k)}, u^{(k)}(t)) = 0, \qquad G_y^{\varphi^{(k)}}(y^{(k)}(T)) = 0,$$

which lead to the equation (3.22). Note that (3.22) is the conjugate system known in the theory of the Pontryagin maximum principle [1]. Thus, we have shown how to obtain the function $p^{(k)}$ specified in (3.21).

To obtain $\Sigma^{(k)}$ consider the relations

$$d^2 R^{\varphi^{(k)}}(t, y^{(k)}(t), u^{(k)}(t)) = \langle \Delta y, R_{yy}^{\varphi^{(k)}}(t, y^{(k)}(t), u^{(k)}(t)) \Delta y \rangle \geqslant 0, \tag{3.26}$$

$$d^2 G^{\varphi^{(k)}}(y^{(k)}(T)) = \langle \Delta y(T), G_{yy}^{\varphi^{(k)}}(y^{(k)}(T)) \rangle \leqslant 0 \tag{3.27}$$

with the corresponding matrices

$$R_{yy}^{\varphi^{(k)}}(t, y^{(k)}(t), u^{(k)}(t)) = \operatorname{diag}\{\delta_1(t), \dots, \delta_n(t)\},$$
$$G_{yy}^{\varphi^{(k)}}(y^{(k)}(T)) = \operatorname{diag}\{\alpha_1, \dots, \alpha_n\},$$

where $\delta_j(t) \geqslant 0$ and $\alpha_j \leqslant 0$, $j = 1, \dots, n$. Based on (3.26), (3.27), we have the following Cauchy problem for the function $\Sigma^{(k)}$:

$$\frac{d\Sigma_{i,j}^{(k)}(t)}{dt} = \frac{\partial^2 f^0(t, y^{(k)}(t), u^{(k)}(t))}{\partial y_i\, \partial y_j} - \sum_{l=1}^{n}\Bigg[\Sigma_{l,i}^{(k)}\frac{\partial f_l(t, y^{(k)}(t), u^{(k)}(t))}{\partial y_j} + \Sigma_{l,j}^{(k)}\frac{\partial f_l(t, y^{(k)}(t), u^{(k)}(t))}{\partial y_i} + p_l(t)\frac{\partial^2 f_l(t, y^{(k)}(t), u^{(k)}(t))}{\partial y_i\, \partial y_j}\Bigg] + \begin{cases} 0, & i \neq j, \\ \delta_i(t) > 0, & i = j, \end{cases} \tag{3.28}$$

$$\Sigma_{i,j}^{(k)}(T) = -\frac{\partial^2 F(y^{(k)}(T))}{\partial y_i\, \partial y_j} - \begin{cases} 0, & i \neq j, \\ \alpha_i > 0, & i = j, \end{cases} \tag{3.29}$$

where $i, j = 1, \dots, n$.

For the problem (3.1)–(3.3) one uses the following iterative procedure, where $v^{(k)} = (y^{(k)}, u^{(k)})$ and $v^{(k+1)} = (y^{(k+1)}, u^{(k+1)})$ are the input and output admissible processes, respectively, at the kth iteration.

1. Define the linear-quadratic function $\varphi^{(k)}$ by finding the functions $p^{(k)}$ and $\Sigma^{(k)}$ as the solutions of the Cauchy problems (3.22) and (3.28), (3.29) with fixed $\delta_i(t)$ and α_i $(i = 1, \dots, n)$, respectively.
2. Find the solution $y^{(k+1)}$ of the Cauchy problem (3.11). Find the control $u^{(k+1)}$ defined by (3.24).

The following theorem is formulated like the theorem in [58].

Theorem 3.3. *For the problem* (3.1)–(3.3) *let the conditions* 1)–3) *in Lemma* 3.1 *be satisfied. Then the method* (3.11), (3.21), (3.22), (3.24), (3.28), (3.29) *provides a sequence of processes* $\{v^{(k)}\} \subset \mathscr{D}$ *such that* $J(v^{(k+1)}) \leqslant J(v^{(k)})$. *Moreover, if* (3.25) *is satisfied, then* $\{v^{(k)}\}$ *is an improving sequence.*

Remark 3.1. If the process $v^{(k)}$ satisfies the Pontryagin maximum principle, then the Krotov method does not improve (see [149], p. 64) this process, including in the case when $v^{(k)}$ is not optimal.

In (3.28) and (3.29) we need the functions $\delta_i(t)$ and the parameters α_i $(i = 1, \dots, n)$ to regulate improvement. The constructions (3.22), (3.28), (3.29) are obtained using the necessary conditions for local extrema, so the role of $\delta_i(t)$ and α_i is to compensate as much as possible for the absence of a search for a global extremum. As noted in [58] and [61], if there is no improvement for the selected $\delta_i(t)$ and α_i $(i = 1, \dots, n)$, then (without changing the index k) we need to increase $\delta_i(t) > 0$, decrease $\alpha_i < 0$, and find the corresponding process $v^{(k+1)}$. Taking into account the symmetry $\Sigma_{i,j}(t) = \Sigma_{j,i}(t)$, we must consider $n(n+1)/2$ equations in (3.28), (3.29).

3.4. Krotov method for bilinear systems.

Lemma 3.2. *For the problem* (3.7), (3.8) *with* $\mathscr{F}(y(T)) = -\langle y(T), My(T)\rangle$, $M \geqslant 0$, *the conjugate system* (3.22), *considered on some process* $v^{(k)} \in \mathscr{D}$, *has the form*

$$\frac{dp^{(k)}(t)}{dt} = -\left(A^{\mathrm{T}}(t) + \sum_{l=1}^{m} B_l^{\mathrm{T}} u_l^{(k)}(t)\right) p^{(k)}(t), \qquad p^{(k)}(T) = 2My^{(k)}(T). \quad (3.30)$$

Lemma 3.3. *In the problem* (3.7), (3.8) *let*

$$\lambda_u > 0 \quad \textit{and} \quad Q = [a_1, b_1] \times \cdots \times [a_m, b_m].$$

Then for (3.12) *with a linear-quadratic function* $\varphi^{(k)}(t, y)$ *of the form* (3.21) *one has*

$$\widetilde{u}_l^{(k)}(t, y) = \begin{cases} a_l, & u_{l,\mathrm{st}}^{(k)}(t, y) < a_l, \\ u_{l,\mathrm{st}}^{(k)}(t, y), & u_{l,\mathrm{st}}^{(k)}(t, y) \in [a_l, b_l], \\ b_l, & u_{l,\mathrm{st}}^{(k)}(t, y) > b_l, \end{cases}$$

$$u_{l,\mathrm{st}}^{(k)}(t, y) = \frac{\langle p^{(k)}(t) + \Sigma^{(k)}(t)\Delta y, B_l(t) y\rangle}{2\lambda_u}, \qquad l = 1, \ldots, m,$$

where the function $p^{(k)}$ *satisfies* (3.30), *and* $\Sigma^{(k)}$ *is defined by* (3.23) *or by* (3.28), (3.29). *Here the* $\widetilde{u}_l(t, y)$ *are continuous with respect to* y.

Let $\varphi^{(k)}$ be considered in the class of linear functions:

$$\varphi^{(k)}(t, y) = \langle p^{(k)}(t), y\rangle.$$

This choice corresponds to the first-order Krotov method. For the maximisation problem (3.9), where

$$G^{\varphi^{(k)}}(y(T)) = -\langle y(T), My(T)\rangle + \langle p^{(k)}(T), T\rangle - \langle p^{(k)}(0), x_0\rangle,$$

the condition $M \geqslant 0$ is important since it provides a concave function $\mathscr{F}(y(T))$.

Consider two cases for λ_u: $\lambda_u = 0$ and $\lambda_u > 0$. The following lemma holds ([147], [150], [151]).

Lemma 3.4. *In the problem* (3.7), (3.8) *let* $m = 1$ *and* $Q = [a, b]$. *Then for* (3.12) *with the linear function* $\varphi^{(k)}(t, y) = \langle p^{(k)}(t), y\rangle$ *one has*:

(a) *if* $\lambda_u = 0$, *then the function* $\widetilde{u}^{(k)}(t, y)$ *is defined by* (3.19), *where*

$$u_{\mathrm{sing}}^{(k)}(t, y) = u^{(k)}(t) + \frac{\langle p^{(k)}(t), \big(A(t)B(t) - B(t)A(t) - dB(t)/dt\big) y\rangle}{\langle p^{(k)}(t), (B(t))^2 y\rangle}; \quad (3.31)$$

(b) *if* $\lambda_u > 0$, *then the function* $\widetilde{u}^{(k)}(t, y)$ *is defined by* (3.20), *where*

$$u_{\mathrm{st}}^{(k)}(t, y) = \frac{\langle p^{(k)}(t), By\rangle}{2\lambda_u}. \quad (3.32)$$

In contrast to the case $\lambda_u = 0$, for $\lambda_u > 0$ the function $\widetilde{u}^{(k)}(t, y)$ is continuous with respect to y. The formula (3.31) is obtained by differentiating the switching function $g^{(k)}(t, y) = \langle p^{(k)}(t), By\rangle$. The function (3.32) is obtained from the condition $\partial H/\partial u = 0$.

4. First-order Krotov method for controlling quantum systems governed by the Schrödinger and Liouville–von Neumann equations

After [62] (Krotov and Kazakov, 1987) and [63] (Krotov, 1989), the next important contribution to adaptation of the Krotov method for optimal quantum control was in [64] (Tannor, Kazakov, and Orlov, 1992) and [65] (Somlói, Kazakov, and Tannor, 1993), which contain an application of the first-order Krotov method (with linear function φ) to optimisation of controls for quantum systems governed by the Schrödinger equation. Based on these and subsequent publications, this section outlines theoretical and numerical results on applications of the first-order Krotov method to quantum systems governed by the Schrödinger and Liouville–von Neumann equations.

4.1. Krotov method for the Schrödinger equation. Let us consider the OCP (2.1)–(2.4). The Pontryagin function for this problem is

$$H(t,q,\psi,u) = 2\,\mathrm{Re}\left\langle q, -\frac{i}{\hbar}\mathbf{H}[u]\psi\right\rangle_{\mathscr{H}} - \lambda_\psi\langle\psi, D(t)\psi\rangle_{\mathscr{H}} - \frac{\lambda_u\|u\|^2}{S(t)}, \tag{4.1}$$

where $\psi, q \in \mathscr{H}$ and $u \in Q \subseteq \mathbb{R}^m$. By analogy with (3.14), the Krotov Lagrangian with linear function $\varphi(t,\psi) = 2\,\mathrm{Re}\langle\chi(t),\psi\rangle$ for this problem is the functional

$$L^\varphi(v) = G^\varphi(\psi(T)) - \int_0^T R^\varphi(t,\psi(t),u(t))\,dt, \qquad v = (\psi,u), \tag{4.2}$$

where

$$G^\varphi = -\langle\psi(T), O\psi(T)\rangle + 2\,\mathrm{Re}\langle\chi(T),\psi(T)\rangle - 2\,\mathrm{Re}\langle\chi(0),\psi(0)\rangle, \tag{4.3}$$

$$R^\varphi = 2\,\mathrm{Re}\left[\left\langle\chi(t), -\frac{i}{\hbar}\mathbf{H}[u]\psi\right\rangle + \left\langle\frac{d\chi(t)}{dt},\psi\right\rangle\right] - \lambda_\psi\langle\psi, D(t)\psi\rangle - \frac{\lambda_u\|u\|^2}{S(t)}. \tag{4.4}$$

Consider the operators $O \geqslant 0$ and $D(t) \geqslant 0$. By analogy with (3.4)–(3.6) and relying on [64], we write down the first-order Krotov method. Consider the following iterative procedure, where $v^{(k)} = (\psi^{(k)}, u^{(k)})$ and $v^{(k+1)} = (\psi^{(k+1)}, u^{(k+1)})$ are the input and output admissible processes, respectively, at the kth iteration of the method.

1. Compute the solution $\chi^{(k)}$ of the Cauchy problem

$$\begin{aligned}\frac{d\chi^{(k)}(t)}{dt} &= -\frac{i}{\hbar}\mathbf{H}[u^{(k)}(t)]\chi^{(k)}(t) + \lambda_\psi D(t)\psi^{(k)}(t),\\ \chi^{(k)}(T) &= O\psi^{(k)}(T)\end{aligned} \tag{4.5}$$

(the system is integrated 'from right to left').

2. Find the solution $\psi^{(k+1)}$ of the Cauchy problem

$$\frac{d\psi^{(k+1)}(t)}{dt} = -\frac{i}{\hbar}\mathbf{H}[\widetilde{u}^{(k)}(t,\psi^{(k+1)}(t))]\psi^{(k+1)}(t), \qquad \psi^{(k+1)}(0) = \psi_0. \tag{4.6}$$

Find the control $u^{(k+1)}$ defined by

$$u^{(k+1)}(t) = \widetilde{u}^{(k)}(t,\psi^{(k+1)}(t)) := \arg\max_{u\in Q} H(t,\chi^{(k)}(t),\psi^{(k+1)}(t),u). \tag{4.7}$$

By analogy with the proof of Theorem 3.1, one can prove the following theorem.

Theorem 4.1. *For the problem (2.1)–(2.4) with operators $O \geqslant 0$ and $D(t) \geqslant 0$ and coefficient $\lambda_\psi \leqslant 0$, the method (4.5)–(4.7) provides a sequence of processes $v^{(k)} \in \mathscr{D}$ such that $J(v^{(k+1)}) \leqslant J(v^{(k)})$. Moreover, if*

$$\int_0^T \max_{u\in Q} H\bigl(t, \chi^{(k)}(t), \psi^{(k)}(t), u\bigr)\,dt \neq \int_0^T H\bigl(t, \chi^{(k)}(t), \psi^{(k)}(t), u^{(k)}(t)\bigr)\,dt,$$

then $\{v^{(k)}\}$ is an improving sequence.

Remark 4.1. Realisation of the Krotov method depends on whether there is a regulariser $\lambda_u \displaystyle\int_0^T \frac{\|u(t)\|^2}{S(t)}\,dt$ in (2.4) ($\lambda_u > 0$), or not ($\lambda_u = 0$). For illustration, let $Q = [a_1, b_1] \times \cdots \times [a_m, b_m]$ in the problem (2.1)–(2.4).

If $\lambda_u = 0$, then for (4.7) consider the function $\widetilde{u}^{(k)}(t, \psi)$ defined by the formula

$$\widetilde{u}_l^{(k)}(t,\psi) = \begin{cases} a_l, & g_l^{(k)}(t,\psi) < 0, \\ u_{l,\mathrm{sing}}^{(k)}(t,\psi) \in [a_l, b_l], & g_l^{(k)}(t,\psi) = 0, \\ b_l, & g_l^{(k)}(t,\psi) > 0, \end{cases}$$

where

$$g_l^{(k)}(t,\psi) = 2\,\mathrm{Re}\left\langle \chi^{(k)}(t), -\frac{i}{\hbar}\mathbf{H}_l\psi \right\rangle = \frac{2}{\hbar}\,\mathrm{Im}\langle \chi^{(k)}(t), \mathbf{H}_l\psi\rangle, \qquad l = 1, \ldots, m,$$

are the components of the switching function. In the case $m = 1$ Krotov's articles [107] and [108] give a formula for $u_{\mathrm{sing}}^{(k)}(t, \psi)$.

If $\lambda_u > 0$, then for (4.7) the function $\widetilde{u}^{(k)}(t, \psi)$ is defined by

$$\widetilde{u}_l^{(k)}(t,\psi) = \begin{cases} a_l, & u_{l,\mathrm{st}}^{(k)}(t,\psi) < a_l, \\ u_{l,\mathrm{st}}^{(k)}(t,\psi), & u_{l,\mathrm{st}}^{(k)}(t,\psi) \in [a_l, b_l], \\ b_l, & u_{l,\mathrm{st}}^{(k)}(t,\psi) > b_l, \end{cases} \tag{4.8}$$

where the functions

$$u_{l,\mathrm{st}}^{(k)}(t,\psi) = \frac{S(t)}{\hbar\lambda_u}\,\mathrm{Im}\langle \chi^{(k)}(t), \mathbf{H}_l\psi\rangle, \qquad l = 1, \ldots, m, \tag{4.9}$$

are obtained from the condition $\partial H/\partial u = 0$.

In the early 1990s the first-order Krotov method, which relates to the conditions $\lambda_u > 0$ and $S(t) \equiv 1$ in (2.4), was considered in [64] (Tannor, Kazakov, and Orlov, 1992) and [65] (Somlói, Kazakov, and Tannor, 1993), and further in the books [60], pp. 253–259 (Krotov, 1996) and [9], § 16.2.2 (Tannor, 2007). In [107] and [108] (Krotov, 2008, 2009) both the cases $\lambda_u = 0$ and $\lambda_u > 0$ were analysed. If $\lambda_u > 0$, then a trade-off between minimisation of $\mathscr{F}$ and regularisation is important.

Another version of the Krotov method is obtained with the following regulariser. Consider the cost functional $J(v)$ with $\lambda_u = 0$ in (2.4) and the following regularised cost functional to be minimised:

$$\widetilde{J}(v, v^{(k)}) = J(v) + \Gamma(\gamma_u, v, v^{(k)}) \to \min, \tag{4.10}$$

$$\Gamma(\gamma_u, v, v^{(k)}) = \gamma_u \int_0^T \frac{\|u(t) - u^{(k)}(t)\|^2}{S(t)}\, dt, \qquad \gamma_u > 0. \tag{4.11}$$

The modified cost functional $\widetilde{J}(v, v^{(k)})$ is different for each iteration of the Krotov method, because this functional depends on the current approximation $v^{(k)}$.

Lemma 4.1. *Suppose that a process $\widehat{v} \in \mathscr{D}$ improves a given process $v^{(k)} \in \mathscr{D}$ for the regularised functional (4.10), (4.11), that is,*

$$\widetilde{J}(\widehat{v}, v^{(k)}) < \widetilde{J}(v^{(k)}, v^{(k)}) = J(v^{(k)}).$$

Then the process $\widehat{v}$ also improves $v^{(k)}$ for the initial functional J, that is,

$$J(\widehat{v}) < J(v^{(k)}).$$

Proof. To prove this lemma, we will argue by contradiction, that is, suppose that the relations

$$J(\widehat{v}) \geqslant J(v^{(k)}) \quad \text{and} \quad \widetilde{J}(\widehat{v}, v^{(k)}) < \widetilde{J}(v^{(k)}, v^{(k)}) = J(v^{(k)}) \tag{4.12}$$

are satisfied simultaneously. Substituting $\widehat{v}$ into (4.10) and (4.11), we have

$$\widetilde{J}(\widehat{v}, v^{(k)}) = J(\widehat{v}) + \widehat{c},$$

where $\widehat{c} = \Gamma(\gamma_u, \widehat{v}, v^{(k)})$ and $J(\widehat{v}) = \widetilde{J}^{(k)}(\widehat{v}) - \widehat{c}$. It is clear that $\widehat{c} > 0$ if $\widehat{u}(t) \not\equiv u^{(k)}(t)$. Based on the assumptions (4.12), we obtain

$$J(\widehat{v}) = \widetilde{J}(\widehat{v}, v^{(k)}) - \widehat{c} \geqslant J(v^{(k)}) > \widetilde{J}(\widehat{v}, v^{(k)}).$$

Thus, we get that $\widetilde{J}(\widehat{v}, v^{(k)}) - \widehat{c} > \widetilde{J}(\widehat{v}, v^{(k)})$, that is, $\widehat{c} < 0$. The latter contradicts the fact that $\widehat{c} > 0$ if $\widehat{u}(t) \not\equiv u^{(k)}(t)$. If $\widehat{u}(t) \equiv u^{(k)}(t)$, then there is no improvement for $v^{(k)}$ in relation to $\widetilde{J}$. We conclude that $J(\widehat{v}) < J(v^{(k)})$. □

In the problem (2.1)–(2.4) let

$$Q = [a_1, b_1] \times \cdots \times [a_m, b_m] \quad \text{and} \quad \lambda_u = 0.$$

Consider the problem of improving the process $v^{(k)} \in \mathscr{D}$. Using (4.10) and (4.11), one obtains the corresponding version of the Krotov method, where the formula (4.8) is used, and the components of the vector $u_{\mathrm{st}}^{(k)}(t, \psi)$ are defined not by (4.9), but as follows:

$$u_{l,\mathrm{st}}^{(k)}(t, \psi) = u_l^{(k)}(t) + \frac{S(t)}{\hbar \gamma_u} \operatorname{Im}\langle \chi^{(k)}(t), \mathbf{H}_l \psi \rangle, \qquad l = 1, \ldots, m, \quad \gamma_u > 0.$$

This formula is obtained from the condition

$$\frac{\partial H}{\partial u_l} = 2\,\mathrm{Re}\left\langle \chi^{(k)}(t), -\frac{i}{\hbar}\mathbf{H}_l\psi \right\rangle - \frac{2\gamma_u(u_l - u_l^{(k)}(t))}{S(t)} = 0.$$

The Krotov method with (4.10) is used to solve OCPs for systems governed by the Schrödinger equation, for example, in [71] (Koch, Palao, Kosloff, and Masnou-Seeuws, 2004). In [66] (Sklarz and Tannor, 2002) the Krotov method combined with such a regulariser was used for an OCP with the Gross–Pitaevskii equation, which will be discussed later in this survey. The first- and second-order versions of the Krotov method have been used in combination with this regularisation in a number of papers since 2002, including, for example, [22], [67], [73], [74], [78], [89], and [94].

The functional (4.10), (4.11) is designed to regulate the distance to the current approximation $u^{(k)}$. Such regularisation is used in the theory of optimal control; see, for example, [146], p. 61 (Srochko, 2000).

4.2. Krotov method for the Liouville–von Neumann equation. The Krotov method has been applied to the solution of OCPs for open quantum systems, including for systems of the form (2.10) ([22], [70], [79], [92], [93], [95], [104]). The Liouville–von Neumann equation (2.9) with a controlled Hamiltonian follows from (2.10) for the dissipator $\mathscr{L}(\rho) \equiv 0$.

Consider the problem (2.3), (2.9), (2.11) with $\lambda_u = 0$ and $\lambda_\rho > 0$. The conjugate system has the form

$$\frac{d\sigma^{(k)}(t)}{dt} = -\frac{i}{\hbar}\big[\mathbf{H}[u^{(k)}(t)], \sigma^{(k)}(t)\big] - \lambda_\rho D(t), \qquad \sigma^{(k)}(T) = \rho_{\mathrm{target}}. \tag{4.13}$$

We briefly describe the application of the first-order Krotov method to this problem. Consider the following iterative process, where $v^{(k)} = (\rho^{(k)}, u^{(k)})$ and $v^{(k+1)} = (\rho^{(k+1)}, u^{(k+1)})$ are the input and output admissible processes, respectively, at the kth iteration of the method.

1. Compute the solution $\sigma^{(k)}$ of the Cauchy problem (4.13).
2. Find the solution $\rho^{(k+1)}$ of the Cauchy problem

$$\frac{d\rho^{(k+1)}(t)}{dt} = -\frac{i}{\hbar}\big[\mathbf{H}[\widetilde{u}^{(k)}(t, \rho^{(k+1)}(t))], \rho^{(k+1)}(t)\big], \quad \rho^{(k+1)}(0) = \rho_0. \tag{4.14}$$

Find the control $u^{(k+1)}$ defined by the formula

$$u_l^{(k+1)}(t) = \widetilde{u}_l^{(k)}(t, \rho^{(k+1)}(t)) := \begin{cases} a_l, & u_{l,\mathrm{st}}^{(k)}(t, \rho^{(k+1)}(t)) < a_l, \\ b_l, & u_{l,\mathrm{st}}^{(k)}(t, \rho^{(k+1)}(t)) > b_l, \\ u_{l,\mathrm{st}}^{(k)}(t, \rho^{(k+1)}(t)), & u_{l,\mathrm{st}}^{(k)}(t, \rho^{(k+1)}(t)) \in [a_l, b_l], \end{cases} \tag{4.15}$$

where

$$u_{l,\mathrm{st}}^{(k)}(t,\rho^{(k+1)}(t)) = u_l^{(k)}(t) + \frac{S(t)}{\gamma_u\hbar}\operatorname{Im}\bigl(\operatorname{Tr}\bigl\{\sigma^{(k)}(t)\bigl(\mathbf{H}_l\rho^{(k+1)}(t) - \rho^{(k+1)}(t)\mathbf{H}_l\bigr)\bigr\}\bigr), \quad (4.16)$$

$l = 1, \dots, m$ and $\gamma_u > 0$.

Theorem 4.2. *For the problem* (2.3), (2.9), (2.11) *with* $\lambda_u = 0$ *and* $\lambda_\rho > 0$, *the method* (4.13)–(4.16) *provides a sequence of processes* $\{v^{(k)}\} \subset \mathscr{D}$ *such that* $J(v^{(k+1)}) \leqslant J(v^{(k)})$. *Moreover, if*

$$\int_0^T \max_{u\in Q} H\bigl(t,\sigma^{(k)}(t),\rho^{(k)}(t),u\bigr)\,dt \neq \int_0^T H\bigl(t,\sigma^{(k)}(t),\rho^{(k)}(t),u^{(k)}(t)\bigr)\,dt,$$

then $\{v^{(k)}\}$ *is an improving sequence.*

4.3. Zhu–Rabitz and Maday–Turinici methods. Their connections with the first-order Krotov method. Consider the OCP (2.1)–(2.4) with

$$O \geqslant 0, \quad m = 1, \quad Q = \mathbb{R}, \quad \lambda_u > 0, \quad S(t) \equiv 1, \quad \lambda_\psi = 0,$$

and the Hamiltonian $\mathbf{H} = \mathbf{H}_0 - \boldsymbol{\mu}u(t)$, where $\boldsymbol{\mu}$ is the dipole moment operator. Taking into account the specifics of this OCP, the authors of [42] (Zhu and Rabitz, 1998) proposed a method for non-local improvements which we call the *Zhu–Rabitz method*.

The Pontryagin function for this problem is

$$H(t,\chi,\psi,u) = 2\operatorname{Re}\Bigl\langle \chi, -\frac{i}{\hbar}\mathbf{H}[u]\psi\Bigr\rangle - \lambda_u u^2.$$

Because H is quadratic in u, the condition $\partial H/\partial u = 0$ gives

$$\widetilde{u}(t,\chi,\psi) = -\frac{1}{\lambda_u\hbar}\operatorname{Im}\langle\chi,\boldsymbol{\mu}\psi\rangle. \quad (4.17)$$

Consider the following iterative process, where $v^{(k)} = (\psi^{(k)}, u^{(k)})$ and $v^{(k+1)} = (\psi^{(k+1)}, u^{(k+1)})$ are the input and output admissible processes, respectively, at the kth iteration.

1. Compute the function $\chi^{(k+1)}$ solving the Cauchy problem

$$\frac{d\chi^{(k+1)}(t)}{dt} = -\frac{i}{\hbar}\bigl(\mathbf{H}_0 - \boldsymbol{\mu}\widetilde{u}(t,\chi^{(k+1)}(t),\psi^{(k)}(t))\bigr)\chi^{(k+1)}(t), \quad (4.18)$$

$$\chi^{(k+1)}(T) = O\psi^{(k)}(T), \quad (4.19)$$

obtained by substituting $\widetilde{u}(t,\chi^{(k+1)}(t),\psi^{(k)}(t))$ into the conjugate system (4.5) in place of $u^{(k)}(t)$ for $\lambda_\psi = 0$.

2. Find the function $\psi^{(k+1)}$ which satisfies the Cauchy problem

$$\frac{d\psi^{(k+1)}(t)}{dt} = -\frac{i}{\hbar}\bigl(\mathbf{H}_0 - \boldsymbol{\mu}\widetilde{u}(t,\chi^{(k+1)}(t),\psi^{(k+1)}(t))\bigr)\psi^{(k+1)}(t), \quad (4.20)$$

$$\psi^{(k+1)}(0) = \psi_0. \quad (4.21)$$

Using the formula (4.17), find the control $u^{(k+1)}$ as

$$u^{(k+1)}(t) = \widetilde{u}\big(t, \chi^{(k+1)}(t), \psi^{(k+1)}(t)\big) := -\frac{1}{\lambda_u \hbar} \operatorname{Im}\langle \chi^{(k+1)}(t), \boldsymbol{\mu}\psi^{(k+1)}(t)\rangle. \tag{4.22}$$

Theorem 4.3. *In the problem* (2.1)–(2.4), *let* $O \geqslant 0$, $m = 1$, $Q = \mathbb{R}$, $\lambda_u > 0$, $S(t) \equiv 1$, $\lambda_\psi = 0$, *and* $\mathbf{H} = \mathbf{H}_0 - \boldsymbol{\mu} u(t)$. *The Zhu–Rabitz method* (4.17)–(4.22) *gives a process* $v^{(k+1)} = (\psi^{(k+1)}, u^{(k+1)}) \in \mathscr{D}$ *such that* $J(v^{(k+1)}) \leqslant J(v^{(k)})$.

The Zhu–Rabitz method (4.17)–(4.22) was presented in [42], where numerical results were also presented showing that this method gives good improvements, and that $J(v)$ is improved more quickly in the first few iterations than in subsequent iterations. In [109] (Sundermann and de Vivie-Riedle, 1999) the Zhu–Rabitz method was applied with the shape function $S(t) = \sin^2(\pi t/T)$.

In [43] (Maday and Turinici, 2003) a general method was proposed which includes as particular cases the first-order Krotov method and the Zhu–Rabitz method. We will call it the *Maday–Turinici method*. It works as follows.

Fix two parameters: $\delta, \eta \in [0, 2]$.

1. Find the solution $\psi^{(k+1)}$ of the Cauchy problem

$$\frac{d\psi^{(k+1)}(t)}{dt} = -\frac{i}{\hbar}\mathbf{H}[\widetilde{u}(t, \psi^{(k+1)}(t); \delta)]\psi^{(k+1)}(t), \qquad \psi^{(k+1)}(0) = \psi_0. \tag{4.23}$$

Find the control $u^{(k+1)}$ defined by

$$u^{(k+1)}(t) = \widetilde{u}(t, \psi^{(k+1)}(t); \delta) := (1-\delta)u^{(k)}(t) - \frac{\delta}{\lambda_u \hbar} \operatorname{Im}\langle \chi^{(k)}(t), \boldsymbol{\mu}\psi^{(k+1)}(t)\rangle, \tag{4.24}$$

where the functions $u^{(k)}$ and $\chi^{(k)}$ are from the previous iteration.

2. Find the function $\chi^{(k+1)}$ as the solution of the Cauchy problem

$$\frac{d\chi^{(k+1)}(t)}{dt} = -\frac{i}{\hbar}\mathbf{H}[\widetilde{u}(t, \chi^{(k+1)}(t); \eta)]\chi^{(k+1)}(t), \qquad \chi^{(k+1)}(T) = O\psi^{(k+1)}(T), \tag{4.25}$$

where

$$\widetilde{u}(t, \chi^{(k+1)}; \eta) := (1-\eta)\widetilde{u}(t, \psi^{(k+1)}(t); \delta) - \frac{\eta}{\lambda_u \hbar} \operatorname{Im}\langle \chi^{(k+1)}, \boldsymbol{\mu}\psi^{(k+1)}(t)\rangle. \tag{4.26}$$

Theorem 4.4. *In the problem* (2.1)–(2.4), *let* $O \geqslant 0$, $m = 1$, $Q = \mathbb{R}$, $\lambda_u > 0$, $S(t) \equiv 1$, $\lambda_\psi = 0$, *and* $\mathbf{H} = \mathbf{H}_0 - \boldsymbol{\mu} u(t)$. *Then for any* $\eta, \delta \in [0, 2]$ *the Maday–Turinici method* (4.23)–(4.26) *gives* $J(v^{(k+1)}) \leqslant J(v^{(k)})$.

The parameters (δ, η) in the Maday–Turinici method (4.23)–(4.26) should be chosen correctly. The choice $(\delta, \eta) = (1, 0)$ determines a version of the first-order Krotov method, and the choice $(\delta, \eta) = (1, 1)$ leads to the Zhu–Rabitz method.

As noted in [43], the Maday–Turinici method can be better than the first-order Krotov method or the Zhu–Rabitz method. The Maday–Turinici method was also developed in the case where a controlled evolution of the density matrix is considered [152] (Ohtsuki, Turinici, and Rabitz, 2004).

Both the Zhu–Rabitz and the Maday–Turinici methods exploit the regulariser $\lambda_u \int_0^T (u(t))^2\,dt$, $\lambda_u > 0$, in the cost functional. This regulariser implies that the Pontryagin function is quadratic in u, which is convenient for maximising it. On other hand, if the original cost functional J has no term of type $\lambda_u \int_0^T (u(t))^2\,dt$, then a trade-off between minimising the terminal and integral parts of J is important.

4.4. Krotov method and other methods in numerical experiments for the controlled Schrödinger and Liouville–von Neumann equations. Based on a number of publications since the early 1990s, we outline applications of the first-order Krotov method and other methods to the problem (2.1)–(2.4) with $\psi(t) \in L^2$ or $\psi(t) \in \mathbb{C}^n$. For the OCP under consideration, gradient methods such as steepest-descent and conjugate-gradient methods have been used in the control space for a long time [68], [153].

Definition 4.1. The following iterative method is called the steepest-descent method in the problem (2.1)–(2.4):

$$u^{(k)}(t;\beta) = u^{(k)}(t) + \beta \frac{\partial H}{\partial u}\big(t, \chi^{(k)}(t), \psi^{(k)}(t), u^{(k)}(t)\big), \qquad \beta > 0, \tag{4.27}$$

$$u^{(k+1)}(t) = u^{(k)}(t;\beta = \widehat{\beta}), \qquad \widehat{\beta} = \arg\min_{\beta>0} J\big(\psi^{(k)}(\,\cdot\,;\beta), u^{(k)}(\,\cdot\,;\beta)\big), \tag{4.28}$$

where $(\psi^{(k)}, u^{(k)})$ is a given process that is to be improved, $H(t,\chi,\psi,u)$ is the Pontryagin function (4.1), and $\psi^{(k)}(\cdot;\beta)$ is the solution of the Cauchy problem (2.1) with the control $u^{(k)}(\,\cdot\,;\beta)$.

In the steepest-descent method, the complexity of a single iteration is essentially determined by a multiple integration of the Schrödinger equation for a family of controls $\{u^{(k)}(\,\cdot\,;\beta),\ \beta > 0\}$, which is necessary in order to find the value $\widehat{\beta}$ that provides a variation of the control with the greatest possible decrease of J in this iteration.

In addition to (2.1)–(2.4), one can specify spectral constraints on the control. For such a problem a modified steepest-descent method was developed in [153] (Gross, Neuhauser, and Rabitz, 1992), where, in contrast to (4.27), the following formula with the Fourier transformation is used:

$$u^{(k+1)}(t;\beta) = u^{(k)}(t) + \frac{\beta}{2\pi} \int_{-\infty}^{\infty} \left[\int_0^T \frac{\partial H}{\partial u}\big(t, \chi^{(k)}(t), \psi^{(k)}(t), u^{(k)}(t)\big) e^{-i\omega t}\,dt \right] g(\omega) e^{i\omega t}\,d\omega,$$

with $\beta > 0$ and

$$g(\omega) = \begin{cases} 1, & \omega_{\min} \leqslant |\omega| \leqslant \omega_{\max}, \\ 0, & |\omega| < \omega_{\min},\ |\omega| > \omega_{\max}. \end{cases}$$

In [154] (2009), this modification of the steepest-descent method was adapted for solution of an OCP for the Liouville–von Neumann equation (2.9).

The article [65] (Somlói, Kazakov, and Tannor, 1993) used the first-order Krotov method for OCPs (2.1)–(2.4) with $\lambda_u > 0$, $S(t) \equiv 1$, and $\lambda_\psi = 0$ to model controlled dissociation of an iodine molecule I_2 by the Schrödinger equation (2.1) with $M = 2$ (two electronic states):

$$\frac{d\psi(t)}{dt} = -\frac{i}{\hbar}\begin{pmatrix} \mathbf{H}_{\mathrm{gr}} & -\mu u(t) \\ -\mu u(t) & \mathbf{H}_{\mathrm{ex}} \end{pmatrix}\psi(t), \qquad \psi(0) = \psi_0, \quad \psi = (\psi_{\mathrm{gr}}, \psi_{\mathrm{ex}})^{\mathrm{T}}.$$

The operators $\mathbf{H}_{\mathrm{gr}}$ and $\mathbf{H}_{\mathrm{ex}}$ and the functions ψ_{gr} and ψ_{ex} describe the ground and excited electronic states, μ means equal dipole operators between the two electronic states of the iodine molecule (in the general case different dipole operators $\mu_{\mathrm{gr,ex}}$ and $\mu_{\mathrm{ex,gr}}$ are considered), and each electronic state has a Morse-type potential

$$V_j = D_{\mathrm{e},j}\{1 - \exp[-\beta_j(r - r_{\mathrm{e},j})]\}^2$$

for certain values of the parameters. Here $D_{\mathrm{e},j}$ is the dissociation energy, r is the nuclear distance between two atoms, $r_{\mathrm{e},j}$ is the equilibrium nuclear distance, and $j \in \{\mathrm{gr}, \mathrm{ex}\}$. It was shown that the Krotov method: (a) can provide macro-steps compared with local improvements produced by the steepest-descent method; (b) does not involve the expensive operation of finding the variational parameter; (c) can improve J at a lower computational cost than the steepest-descent method. Figures 4 and 6 in [65], p. 92, show the dependence of the maximised probability of dissociation on the number of Cauchy problems solved (graphs are plotted for up to 20 Cauchy problems) and demonstrate the advantage of the Krotov method over the gradient method. The first few iterations of the first-order Krotov method give the main contribution and then the rate of change of the values of J decreases (a target population of 99% at time T is obtained). The Husimi transform was applied for the analysis of the optimised control in the joint time-frequency domain.

The paper [68] (Szakács, Amstrup, Gross, Kosloff, Rabitz, and Lörincz, 1994) considers the Schrödinger equation for two electronic states for modelling controlled blocking of the molecular bond for caesium iodide (CsI). The first-order Krotov method is used together with the Fletcher–Reeves form of the conjugate-gradient method in a function space: first with the Krotov method, and then with a switch to the Fletcher–Reeves method, starting with the result of the Krotov method.

In [42] (Zhu and Rabitz, 1998) the problem (2.1)–(2.4) was considered with the Hilbert space $\mathscr{H} = L^2(\mathbb{R})$, the observable

$$O(x) = (\gamma_0\sqrt{\pi}\,)\exp(-\gamma_0^2(x - x')^2),$$

$S(t) \equiv 1$, $m = 1$, $Q = \mathbb{R}$ and $\lambda_\psi = 0$. The aim was to localise the wavepacket at a given location x' (its value was taken equal to 2.5) according to the operator O, with the Morse potential

$$V(x) = D_0(e^{-\beta(x-x_0)} - 1)^2 - D_0$$

of the O–H bond, where D_0, β, and x_0 have certain numerical values. Numerical results show that the Zhu–Rabitz method in the first few iterations converges to

about 80% of its converged value. The article [43] (Maday and Turinici, 2003) considers the same OCP for two values of x' (2.5 and 1.821) and presents comparative results computed for three cases of the Madey–Turinici method. The pairs $(\delta, \eta) = (1, 1)$ and $(\delta, \eta) = (1, 0)$ in the method represent the Zhu–Rabitz method and the first-order Krotov method, respectively. The third case is $(\delta, \eta) = (2, 0)$. Figures 2 and 3 in [43] show the first 10 iterations of computations for both values of x'. According to these figures, the case $(\delta, \eta) = (1, 0)$ is slower than or almost the same speed as the case $(\delta, \eta) = (2, 0)$, and both these cases are better than the case $(\delta, \eta) = (1, 1)$.

In [69] (Sola, Santamaria, and Tannor, 1998), an OCP for the Schrödinger equation with the Morse potential and with several energy levels was considered, and multi-photon excitations were investigated. Figure 1 in [69] shows the comparative results and the better performance of the first-order Krotov method (the first 40 iterations were considered) versus three gradient methods, including the conjugate-gradient method, in the problem of maximising the probability of selecting the target state.

In [49] (Ho and Rabitz, 2010) the problem of maximising the mean

$$\langle O_T \rangle = \langle \psi(T), O_T \psi(T) \rangle$$

with respect to the Schrödinger equation was considered and the TBQCP method was proposed. For the target observable O_T an explicit time-dependent positive-semidefinite Hermitian operator $O(t)$ was considered which is a dynamical invariant (that is, the total derivative $dO(t)/dt$ is equal to zero) and satisfies the condition $O(T) = O_T$. For a certain OCP connected with the Morse potential, Figs. 2 and 4–7 in [49] show that the Ho–Rabitz TBQCP method converges faster than the first-order Krotov method.

The paper [67] (Palao, Kosloff, and Koch, 2008) considers the problem (2.1)–(2.4) with $\lambda_\psi \geqslant 0$ for a model of vibrations in a rubidium molecule Rb_2, where three electronic states are taken. The goal is to transfer population initially at level $v = 0$ of the electronic ground state, to level $v = 1$ of the same electronic state at time T, by means of Raman-like transitions involving levels in the ${}^1\Sigma_u^+$ excited state, but without populating levels in the upper electronic state ${}^1\Pi_g$ at any time. Figure 2 in [67] shows the values of the maximised normalised cost functional J_{norm} and the average population in the allowed subspace I_P, as functions of the number of iterations both with and without this state constraint. In both cases, the first-order Krotov method, which uses the regularisation (4.10), gives a population transfer greater than 99.9%. However, as shown in Figs. 2 and 3 in [67], here the state constraint must be used to avoid populating a forbidden subspace, and the Krotov method successfully solves the problem with this constraint. The cost of using the constraint is a significant increase in the number of iterations of the Krotov method required in comparison with the case without the constraint: 500 versus 17 iterations.

The paper [75] (Kumar, Malinovskaya, and Malinovsky, 2011) considers the Schrödinger equation which describes a three-level Λ-system controlled by a pump field and a Stokes field. The paper presents a comparison of the results computed using the first-order Krotov method, the Zhu–Rabitz method, and the conjugate-gradient method working in the function space for two OCPs: first, for a complete

population transfer, and second, to maximise the coherence between two given energy levels (the first and the third). The paper shows that all the three methods are able to find solutions of these OCPs, including for the cases with $\lambda_\psi > 0$ (state constraints).

Now we consider results on quantum control which were determined by solving OCPs with real-valued states which represent the real and imaginary parts of a complex-valued $\psi(t)$.

The papers [111] (Boussaïd, Caponigro, and Chambrion, 2012) and [77] (Trushkova, 2013) consider the Schrödinger equation with states in $L^2(\Omega;\mathbb{C})$ describing the rotation of a planar molecule, and an approximate system with states in $\mathbb{C}^n$, $n = 22$. This leads to an OCP (2.7), (2.8) with real-valued states $y(t) \in \mathbb{R}^{44}$, where

$$y_j(t) = \operatorname{Re} z_j(t) \quad \text{and} \quad y_{2j}(t) = \operatorname{Im} z_j(t), \quad j = 1, \dots, n.$$

For this OCP the paper [77] uses the Krotov method with $\varphi^{(k)}(t, y)$ obtained from the discrete analogue of the Cauchy problem

$$H\left(t, \frac{\partial \varphi^{(k)}(t,y)}{\partial y}, y, u^{(k)}(t)\right) + \frac{\partial \varphi^{(k)}(t,y)}{\partial t} = 0, \qquad \varphi^{(k)}(T, y) = -F(y),$$

considered in [155] (Trushkova, 2011). The control is restricted by the condition $|u(t)| \leqslant 1/3$ in accordance with the operation $\arg\max_u$ in (3.12). Based on the data in [77], Fig. 1 shows the logarithmic dependence of J on the number of iterations. For the initial iteration $J = 2$, for the fourth iteration $J \approx 1.6 \cdot 10^{-5}$, and for the tenth iteration $J \approx 5.1 \cdot 10^{-6}$.

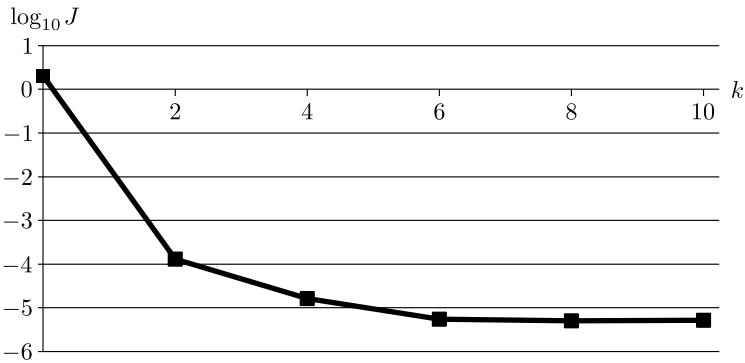

Figure 1. Illustration of the Krotov method in the OCP for rotation of a molecule.

In [151] (Krotov, Morzhin, and Trushkova, 2013), a method which incorporates the first-order Krotov method was proposed for the problem (3.7), (3.8) with

$$\mathscr{F}(y(T)) = -\langle y(T), My(T)\rangle, \qquad M \geqslant 0$$

(and also for a more general class of OCPs). The idea was to consider a generalised OCP which allows *pulse control* and a *discontinuous state function* $y(t)$. The pulse

control combines the usual piecewise continuous control function $u(t)$ and pulse actions used at certain instants of time. At each such time the system's trajectory is described in the state space $\mathbb{R}^n$ using some special dynamical system. By matching the solution $y(t)$ of the initial system, which is integrated between the times of the pulse actions, with the solutions of the special system, which is considered at these times, we obtain a trajectory which is continuous in the state space. Successive improvements of the control are constructed for the generalised OCP. After solving this OCP, its solution can be approximated by processes which are admissible in the initial OCP. Numerical results are provided for an OCP for a quantum system defined by the Schrödinger equation (2.1) with the Landau–Zener Hamiltonian and states $\psi(t) \in \mathbb{C}^2$. For this OCP, its equivalent problem with states $y(t) \in \mathbb{R}^4$ is considered. Table 4 in [151] shows a fast decrease in the cost functional in the generalised OCP.

5. Krotov method for controlling unitary dynamics and ensembles of solutions of the Schrödinger equation

5.1. Second-order Krotov method with constraints on quantum states. In [66] (Sklarz and Tannor, 2002) and [89] (Reich, Ndong, and Koch, 2012) the second-order Krotov method, which uses (3.23) to define $\Sigma^{(k)}$, was developed for OCPs for quantum systems, including for modelling the control of the dynamics of a Bose–Einstein condensate.

Consider the problem (2.19), (2.20), (2.3) (control of the ensemble of solutions of the Schrödinger equation) for $\lambda_\psi \geqslant 0$ without spectral constraints. The Pontryagin function for the regularised cost functional $\widetilde{J}(v, v^{(k)})$ (4.10), (4.11) is

$$H(t, \{q_j\}, \{\psi_j\}, u) = -2\,\mathrm{Re} \sum_{j=1}^{n} \left\langle q_j, \frac{i}{\hbar} \mathbf{H}[u] \psi_j \right\rangle - \frac{\gamma_u \|u - u^{(k)}(t)\|^2}{S(t)},$$

where $\psi_j, q_j \in \mathscr{H}$. We describe the method based on [89] (Reich, Ndong, and Koch, 2012) and [90] (Goerz, Gualdi, Reich, Koch, Motzoi, Whaley, Vala, Müller, Montangero, and Calarco, 2015).

Consider the set of n conjugate systems

$$\frac{d\chi_j^{(k)}(t)}{dt} = -\frac{i}{\hbar} \mathbf{H}[u^{(k)}(t)] \chi_j^{(k)}(t) + \lambda_\psi D(t) \psi_j^{(k)}(t), \tag{5.1}$$

$$\chi_j^{(k)}(T) = -\frac{\partial}{\partial \psi_j} \mathscr{F}\big(\{\psi_j^{(k)}(T)\}_{j=1,\ldots,n}\big), \qquad j = 1, \ldots, n. \tag{5.2}$$

For the problem (2.20), (2.19), (2.3), consider the following iterative procedure, where $v^{(k)} = \big(\{\psi_j^{(k)}\}, u^{(k)}\big)$ and $v^{(k+1)} = \big(\{\psi_j^{(k+1)}\}, u^{(k+1)}\big)$ are the input and output admissible processes, respectively, at the kth iteration of the method.

1. Compute the solutions $\chi_j^{(k)}$, $j = 1, \ldots, n$, of the n Cauchy problems (5.1), (5.2).

2. Find the solutions $\psi_j^{(k+1)}$, $j = 1, \dots, n$, of the n Cauchy problems

$$\begin{aligned} \frac{d\psi_j^{(k+1)}(t)}{dt} &= -\frac{i}{\hbar}\mathbf{H}\big[\widetilde{u}^{(k)}\big(t, \{\psi_j^{(k+1)}(t)\}_{j=1,\dots,n}\big)\big]\psi_j^{(k+1)}(t), \\ \psi_j^{(k+1)}(0) &= \psi_{j,0}. \end{aligned} \tag{5.3}$$

Find the control $u^{(k+1)}$ defined by

$$\begin{aligned} u^{(k+1)}(t) &= \widetilde{u}^{(k)}\big(t, \{\psi_j^{(k+1)}(t)\}_{j=1,\dots,n}\big) \\ &:= \arg\max_{u\in Q} H\bigg(t, \Big\{\chi_j^{(k)}(t) + \frac{1}{2}\Sigma^{(k)}(t)\big(\psi_j^{(k+1)}(t) - \psi_j^{(k)}(t)\big)\Big\}, \\ &\qquad\qquad \{\psi_j^{(k+1)}(t)\}_{j=1,\dots,n}, u\bigg), \end{aligned} \tag{5.4}$$

where the function $\Sigma^{(k)}$ is defined in (3.23) with some $\alpha, \beta < 0$ and $\gamma > 0$.

Theorem 5.1. *For the OCP* (2.20), (2.19), (2.3) *consider the problem of improving the process* $v^{(k)} = \big(\{\psi_j^{(k)}\}_{j=1,\dots,n}, u\big) \in \mathscr{D}$. *Then the method* (5.1)–(5.4) *provides a process* $v^{(k+1)}$ *such that* $J(v^{(k+1)}) \leqslant J(v^{(k)})$.

The article [89] (Reich, Ndong, and Koch, 2012) provides numerical results obtained from the second-order Krotov method for an OCP with the terminant $\mathscr{F}\big(\{\psi_j(T)\}_{j=1,\dots,n}\big)$ (2.22) which is a polynomial of the eighth degree with respect to the ψ_j (see [88], pp. 7–8). As shown in Fig. 1 in [89], the efficiency of the Krotov method depends substantially on γ_u.

5.2. Krotov method with constraints on the control spectrum. It can happen that the control spectrum contains components which are not desirable for practical implementation. Following [76] (Palao, Reich, and Koch, 2013), we describe a modification of the Krotov method for the problem (2.19), (2.20), (2.3) with $\lambda_\psi = 0$, $Q = \mathbb{R}$, and spectral constraints on the control.

At the kth iteration, consider the process $v^{(k)} \in \mathscr{D}$ and the following functional which takes into account both the regulariser (4.10), (4.11) and the frequencies ω_m, $m = 1, \dots, M$, for which filtration is carried out:

$$\begin{aligned} J_{\mathrm{spec}}(v; v^{(k)}) = \int_0^T \bigg[\gamma_u \frac{(u(t) - u^{(k)}(t))^2}{S(t)} \\ + \frac{1}{2\pi}\int_0^T \big(u(t) - u^{(k)}(t)\big)K(t-t')\big(u(t') - u^{(k)}(t')\big)\,dt'\bigg]\,dt. \end{aligned} \tag{5.5}$$

Here the Gaussian kernel has the form

$$K(t-t') = \sum_{m=1}^{M} \lambda_{\mathrm{spec},m}\sqrt{2\pi\sigma_m^2}\,\cos[\omega_m(t-t')]\exp\bigg[-\frac{\sigma_l^2(t-t')^2}{2}\bigg]. \tag{5.6}$$

The values of γ_u and the quantities $\lambda_{\mathrm{spec},m}$ and σ_m $(m = 1, \dots, M)$ are given, and $S(t)$ is some shape function.

Setting $t = t'$, we have

$$K(0) = \sum_{m=1}^{M} \lambda_{\mathrm{spec},m} \sqrt{2\pi\sigma_m^2}\,.$$

The Fourier transform of $K(t-t')$ is

$$\overline{K}(\omega) = \sum_{m=1}^{M} \frac{\lambda_{\mathrm{spec},m}}{2} \left[\exp\left(-\frac{(\omega-\omega_m)^2}{2\sigma_m^2} \right) + \exp\left(-\frac{(\omega+\omega_m)^2}{2\sigma_m^2} \right) \right].$$

Let the Hamiltonian $\mathbf{H}$ be linear in u. Consider the following iterative formula:[7]

$$\begin{aligned} u^{(k+1)}(t) &= \widetilde{u}^{(k)}\big(t, \{\psi_j^{(k+1)}(t), j = 1, \dots, n\}\big) \\ &:= u^{(k)}(t) + \frac{S(t)}{\gamma_u \hbar} \operatorname{Im}\Bigg[\sum_{j=1}^{n} \Big\langle \chi_j^{(k)}(t), \frac{\partial \mathbf{H}}{\partial u} \psi_j^{(k+1)} \Big\rangle \\ &\quad + \frac{1}{2} \sum_{j=1}^{n} \Big\langle \big(\psi_j^{(k+1)}(t) - \psi_j^{(k)}(t)\big), \Sigma^{(k)}(t) \frac{\partial \mathbf{H}}{\partial u} \big(\psi_j^{(k+1)}(t) - \psi_j^{(k)}(t)\big) \Big\rangle \Bigg] \\ &\quad - \sum_{m=1}^{M} \frac{\lambda_{\mathrm{spec},m} S(t)}{2\pi\gamma_u} \sqrt{2\pi\sigma_m^2} \int_0^T \cos[\omega_m(t-t')] \\ &\quad \times \exp\left[-\frac{\sigma_m^2 (t-t')^2}{2} \right] \big(u(t') - u^{(k)}(t')\big)\, dt', \qquad k \geqslant 0, \end{aligned} \tag{5.7}$$

where, at the kth iteration, $u^{(k)}$ and $u^{(k+1)}$ are the current and the next approximations, respectively, $\psi_j^{(k+1)}$, $j = 1, \dots, n$, are the solutions of the Cauchy problems (2.19) with the control

$$u(t) = \widetilde{u}^{(k)}\big(t, \{\psi_j^{(k+1)}(t), j = 1, \dots, n\}\big),$$

and $\chi_j^{(k)}$, $j = 1, \dots, n$, are the solutions of the Cauchy problems (5.1), (5.2) with $\lambda_\psi = 0$.

Due to the linearity of $\mathbf{H}[u]$ with respect to u, (5.7) can be represented as a Fredholm integral equation for the increment $\Delta u = u^{(k+1)} - u^{(k)}$:

$$\Delta u(t) = I(t) + \beta \int_0^T \mathscr{K}(t, t') \Delta u(t')\, dt', \tag{5.8}$$

[7] In (5.7) there is a minus sign before the last summand, in contrast to the corresponding formulae (8), (11) in [76]. At the same time, the kernel $K(t-t')$ in the formula (6b) in [76] contains a minus sign before the summation sign, which is absent in (5.6). Thus, these minus signs compensate each other.

where

$$I(t) = \frac{S(t)}{\gamma_u \hbar} \operatorname{Im}\Bigg[\sum_{j=1}^{n} \Big\langle \chi_j^{(k)}(t), \frac{\partial \mathbf{H}}{\partial u} \psi_j^{(k+1)}(t) \Big\rangle + \frac{1}{2} \sum_{j=1}^{n} \Big\langle \big(\psi_j^{(k+1)}(t) - \psi_j^{(k)}(t)\big), \Sigma^{(k)}(t) \frac{\partial \mathbf{H}}{\partial u} \big(\psi_j^{(k+1)}(t) - \psi_j^{(k)}(t)\big) \Big\rangle \Bigg], \tag{5.9}$$

$$\mathscr{K}(t,t') = -\sum_{l=1}^{L} \frac{\lambda_{\mathrm{spec},l} S(t)}{2\pi\gamma_u} \sqrt{2\pi\sigma_l^2}\, \cos[\omega_l(t-t')] \exp\Big[-\frac{\sigma_l^2 (t-t')^2}{2}\Big]. \tag{5.10}$$

The formula (5.9) includes the unknown functions $\psi_j^{(k+1)}$, $j = 1, \ldots, n$, corresponding to the desired control $u^{(k+1)}$ for which the spectral constraints are given. In [76] the following approach for solving (5.8)–(5.10) was proposed:

1) set the function $\Sigma^{(k)} \equiv 0$;
2) find an improving process

$$\widehat{v}^{(k+1)} = \big(\{\widehat{\psi}_j^{(k+1)}\}_{j=1}^{n}, \widehat{u}^{(k+1)}\big)$$

for the current process $v^{(k)}$ without spectral constraints on the control (in (5.6) set all $\lambda_{\mathrm{spec},l} = 0$);
3) substitute the computed functions $\widehat{\psi}_j^{(k+1)}$, $j = 1, \ldots, n$, into (5.9) in place of the unknown functions $\psi_j^{(k+1)}$, $j = 1, \ldots, n$, to obtain an approximation $\widehat{I}$ for I;
4) solve the equation

$$\Delta u(t) = \widehat{I}(t) + \beta \int_0^T \mathscr{K}(t,t') \Delta u(t')\, dt' \tag{5.11}$$

using the degenerate-kernel method from the theory of integral equations [156] as described in [76], p. 3.

The need to solve the Fredholm equation (5.8) or its simplified form (5.11) complicates each iteration of the Krotov method.

5.3. Maday–Turinici and Krotov methods with modified quality criteria and smoothing of the control. In [99], [100] (Maximov, Nielsen, Salomon, Tošner, and Turinici, 2008, 2010), and also in [110] (Schirmer and de Fouquieres, 2011), the Maday–Turinici method was applied to the OCP (2.3), (2.12), (2.14), for the generation of a target unitary operator.

It is not correct to use the first-order Krotov method or the Maday–Turinici method for every terminant (2.15)–(2.18). Generalising the regularised terminant as proposed in [99], we consider the following regularised cost functional to be minimised:

$$\widetilde{J}_X(U,u;M) = \mathscr{F}_X(U(T)) - \operatorname{Tr}\{U^\dagger(T) M U(T)\} + \lambda_u \int_0^T \frac{\|u(t)\|^2}{S(t)}\, dt, \tag{5.12}$$

where $M \geqslant 0$ is some symmetric matrix with real entries.

In (5.12), consider

$$\mathscr{F}_O(U(T)) = -\operatorname{Tr}\{OU(T)\rho_0 U^\dagger(T)\}$$

defined by (2.16), and $Q = \mathbb{R}^m$. Based on [99], we describe the kth iteration of the Maday–Turinici method using (5.12). Let $\delta, \eta \in [0, 2]$.

1. Compute the solution $U^{(k+1)}$ of the Cauchy problem

$$\frac{dU^{(k+1)}(t)}{dt} = -\frac{i}{\hbar}\mathbf{H}[\widetilde{u}(t, B^{(k)}(t); \delta)]U^{(k+1)}(t), \qquad U^{(k+1)}(0) = \mathbb{I}. \tag{5.13}$$

Find the control $u^{(k+1)}$ defined by the formula

$$\begin{aligned} u_l^{(k+1)}(t) &= \widetilde{u}_l(t, U^{(k+1)}(t); \delta) \\ &:= (1-\delta)u_l^{(k)}(t) + \frac{\delta}{\lambda_u \hbar}\operatorname{Im}\big\langle B^{(k)}(t), \mathbf{H}_l U^{(k+1)}(t)\big\rangle, \end{aligned} \tag{5.14}$$

where $l = 1, \dots, m$ and the functions $u_l^{(k)}$ and $B^{(k)}$ are from the previous iteration.

2. Find the function $B^{(k+1)}$ as the solution of the Cauchy problem

$$\frac{dB^{(k+1)}(t)}{dt} = -\frac{i}{\hbar}\mathbf{H}[\widetilde{u}(t, B^{(k+1)}(t); \eta)]B^{(k+1)}(t), \tag{5.15}$$

$$B^{(k+1)}(T) = OU^{(k+1)}(T)\rho_0 + U^{(k+1)}(T)M, \tag{5.16}$$

where

$$\begin{aligned} \widetilde{u}_l(t, B^{(k+1)}; \eta) &:= (1-\eta)\widetilde{u}_l(t, U^{(k+1)}(t); \delta) \\ &\quad + \frac{\eta}{\lambda_u \hbar}\operatorname{Im}\big\langle B^{(k+1)}, \mathbf{H}_l U^{(k+1)}(t)\big\rangle, \qquad l = 1, \dots, m. \end{aligned} \tag{5.17}$$

In [100] only the case $(\delta, \eta) = (1, 0)$ was considered, where $M = \kappa\mathbb{I}$, $\kappa > 0$. This case corresponds to the first-order Krotov method with regularisations with respect to u and $U(T)$. In addition, smoothing for the computed improving control $u^{(k+1)}$ was proposed. The vector function $u^{(k+1)}_{\text{smooth}}(\,\cdot\,; \alpha)$ with the components

$$u^{(k+1)}_{\text{smooth},l}(\,\cdot\,; \alpha) = (1-\alpha)u_l^{(k+1)} + \alpha F\big(u_l^{(k+1)}\big), \qquad l = 1, \dots, m,$$

was constructed for some $\alpha \in [0, 1]$, where F is a frequency filter, for example, with direct and inverse Fourier transformations. Suppose that $u^{(k+1)}$ for $\alpha = 0$ provides an improvement. Then starting from $\alpha = 1$ and decreasing α, one can search for a smoothed control which also gives an improvement for $(U^{(k)}, u^{(k)})$. This modification is called the *smooth Krotov method* (using the terminology in [100]). Searching for a suitable $\alpha \in [0, 1]$ increases the complexity of each iteration.

5.4. Applications to numerical experiments for controlling unitary dynamics. The articles [67], [76], [80], [81], and [88]–[91] consider OCPs for ensembles of solutions $\{\psi_j(t), j = 1, \dots, n\}$ of the Schrödinger equation using the first- and second-order Krotov methods, including constraints on the states $\{\psi_j(t)\}$ and on the control spectrum.

As noted in [81] (Palao and Kosloff, 2003), there are important questions:

(a) on the existence of a solution to a problem of type (2.19), (2.20), (2.3) taking into account the feasibility of the computed controls in practice and the scalability with increasing n;

(b) on the complexity of computations for finding such a control.

In [89], pp. 7, 8 (Reich, Ndong, and Koch, 2012), Fig. 1 illustrates the first 40 iterations of the first- and second-order versions of the Krotov method which were applied to an OCP with terminant being a polynomial of the eighth degree with respect to $\{\psi_j\}$. The iterative process is sensitive to γ_u in the regularisation of type (4.10), (4.11) and also to the specification of $\Sigma^{(k)}$ in (3.23). In particular, for $\gamma_u = 0.133$ the first-order Krotov method computes fast improvements in the first two iterations, but after that the method gives degraded values of $\mathscr{F}$. For the same γ_u, 40 iterations of the second-order Krotov method give values of the terminant equal to only about 0.01. For $\gamma_u = 0.4$, both the first- and second-order versions of the Krotov method show sufficiently good results: the terminant reaches nearly 10^{-5}. This indicates the importance of using the second-order Krotov method together with an appropriate γ_u for such non-linear terminants.

In [91] (Goerz, Whaley, and Koch, 2015) a multistage optimisation scheme was discussed where in the first stage a reduction of an OCP to the problem of minimising the corresponding function $\widetilde{\mathscr{F}}$ is performed, and after that the solution of the parametrised problem is taken as an initial approximation for the Krotov method.

Consider control parametrisation using trigonometric functions. In addition to the Krotov method, the articles [90], [91], and [93] use an approach based, first, on considering controls in some class of trigonometric functions parametrised by a certain parameter set, and second, on reducing an OCP to minimisation of the corresponding function $\widetilde{\mathscr{F}}$ of these parameters. The CRAB method [41] uses the following parametrisation:

$$u(t) = u_{\text{guess}}(t)\biggl(1 + S(t)\sum_{j=1}^{N}\bigl(a_j \sin(\omega_j t) + b_j \cos(\omega_j t)\bigr)\biggr), \tag{5.18}$$

where $u_{\text{guess}}(t)$ is some initial approximation. This formula uses the Fourier basis to reflect the physical nature of the control. The corresponding objective function is $\widetilde{\mathscr{F}}(a_j, b_j, \omega_j \mid j = 1, \ldots, N)$. To reduce the dimension of the problem, it was proposed to consider $\omega_j = 2\pi j(1 + r_j)/T$, where $r_j \in [-0.5, 0.5]$ is a random number chosen with the uniform distribution. In [90], [91], and [93], the Nelder–Mead method was applied to minimise the objective function. The Krotov method requires differentiability of $\mathscr{F}$, but the approach with reduction of an OCP to the problem of minimising the corresponding function $\widetilde{\mathscr{F}}$ by the Nelder–Mead method does not require one to find the gradient $\nabla\widetilde{\mathscr{F}}$.

Use of (5.18) narrows the search space in an OCP. However, an appropriate parametrisation can be useful for obtaining analytical formulae for the control, which can then be improved using the Krotov method. As noted in [91], in the case of a non-linear terminant $\mathscr{F}$ it is possible to have a situation when the Krotov method, considered for some given initial approximation, encounters a 'plateau' and is insufficiently effective in the sense that even several thousand iterations can give insufficient results. In this case some pre-optimisation is useful, with

reduction of the OCP to a corresponding finite-dimensional optimisation followed by an application of the Nelder–Mead method.

In [67] (Palao, Kosloff, and Koch, 2008) and [76] (Palao, Reich, and Koch, 2013) the Krotov method is used with constraints on the states $\{\psi_j(t)\}$ and on the spectrum of u taken into account. The quantum Fourier transform (W_{QFT} gate) based on a unitary transformation in the model with three electronic states for the molecule Rb_2 is considered. The constraint on states reflects the fact that the upper electronic state $^1\Pi_g$) is forbidden. Optimisation of the control for a unitary transformation under state constraints is successfully attained using the first-order Krotov method. Figure 6 in [67] shows that 50 iterations of the method are not sufficient to avoid populating a forbidden subspace, but 500 iterations of the method do provide almost zero population in the forbidden subspace throughout the course of time. State constraints lead to inhomogeneous equations (5.1), and in this connection we note the article [157] (Ndong, Tal-Ezer, Kosloff, and Koch, 2009) on a Chebychev propagator for such equations. The effectiveness of the Krotov method (5.5)–(5.11), which provides improvements of the control with filtration of its spectrum, is illustrated in [76].

A modification of the Krotov method was used to estimate the time and gate complexity of the generation of multi-qubit unitary operators [85] (Koike and Okudaira, 2010). For the target operators, the unitary operator W_{QFT} realising the N-qubit quantum Fourier transform and a certain unitary operator W_{f} which does not have any apparent symmetry were considered. The operator W_{QFT} has a polynomial gate complexity, and by construction, W_{f} is expected to have exponential complexity. The quality $\mathscr{F}_W$ is defined by (2.15), where $W = W_{\mathrm{QFT}}$ or $W = W_{\mathrm{f}}$. The modified Krotov scheme was implemented to obtain solutions with regard to optimisation of both quality and time for quantum calculations. As a result, the time complexity for W_{QFT} was found to be linear in the number of qubits, while the time complexity for W_{f} was exponential.

The article [99] (Maximov, Tošner, and Nielsen, 2008) describes a method incorporating the Maday–Turinici method and regularisation for $U(T)$ (see (5.12)–(5.17) with $M = \kappa\mathbb{I}$ and $\kappa > 0$), and applications of this method to OCPs connected with nuclear magnetic resonance and dynamic nuclear polarisation. Various values of the parameters $\delta, \eta \in [0, 2]$ were used. For a model of two-spin Hermitian coherence transfer, Fig. 4 in [99] illustrates the results of this method for the same initial approximation $u^{(0)}$ and different $\delta, \eta \in [0, 2]$. This figure shows how many iterations are needed to produce a change in the cost functional of less than 10^{-4}. Very similar results can be achieved in 40–50 iterations for certain pairs (δ, η), and in 100–200 iterations for certain other pairs, and so on. For example, for $(\delta, \eta) = (1, 0)$, which corresponds to a version of the Krotov method, the number of iterations is 60. For a model of coherence transfer, Fig. 13 in [99] shows that increasing the number of spins gives a faster increase in the processor time for GRAPE than for the Maday–Turinici method combined with the regularised terminant. In the case of five spins, the difference in complexity amounts to a factor of 3.8. However, note that the effectiveness of the Maday–Turinici method depends on the parameters λ_u, δ, η, and also on κ.

6. Second-order Krotov method for controlling a Bose–Einstein condensate governed by the Gross–Pitaevskii equation

6.1. Non-linear dynamics and the second-order Krotov method. In the problem (2.1)–(2.4) the dynamical equation (Schrödinger equation) is linear with respect to ψ, and the terminant $\mathscr{F}$ is defined for a positive-semidefinite operator O. As noted in Section 4, under these conditions one can use the Krotov method with the linear function

$$\varphi(t,\psi) = 2\,\mathrm{Re}\langle\chi(t),\psi\rangle,$$

the Zhu–Rabitz method, and the Maday–Turinici method. In the problem (2.23), (2.3), (2.30) the dynamical equation (Gross–Pitaevskii equation) is non-linear with respect to ψ. Therefore, the second-order Krotov method with linear-quadratic function φ is needed.

In [66] (Sklarz and Tannor, 2002), an important step was taken: the second-order Krotov method was extended to OCPs with a controlled Gross–Pitaevskii equation. The article [40] (Jäger, Reich, Goerz, Koch, and Hohenester, 2014) is also devoted to applying the Krotov method to optimisation of controls for a Bose–Einstein condensate.

Consider OCPs for the Gross–Pitaevskii equation (2.23) together with the cost functional $J(v)$ given in (2.30) with $\lambda_u = 0$ and $\lambda_{du} = 0$. For each iteration of the Krotov method, the regularised cost functional $\widetilde{J}(v, v^{(k)})$ given in (4.10) is used. The Pontryagin function is

$$H(t,q,\psi,u) = 2\,\mathrm{Re}\left\langle q, -\frac{i}{\hbar}\bigl(K + V(\,\cdot\,,u) + \kappa|\psi|^2\bigr)\psi\right\rangle - \gamma_u\frac{\|u-u^{(k)}(t)\|^2}{S(t)}, \quad (6.1)$$

where $q,\psi \in \mathscr{H}$, $u \in Q$, and $\lambda_u > 0$. We give the following definition by analogy with Definition 3.5 (the formula (3.21)).

Definition 6.1. The function $\varphi(t,\psi)$ is said to be linear-quadratic if it has the form

$$\begin{aligned}\varphi(t,\psi) = {}& \langle\chi(t),\psi\rangle_{L^2} + \langle\psi,\chi(t)\rangle_{L^2} \\ &+ \frac{1}{2}\bigl\langle\psi-\psi^{(k)}(t), \Sigma(t)(\psi-\psi^{(k)}(t))\bigr\rangle_{L^2},\end{aligned} \quad (6.2)$$

where $\psi^{(k)}$ is the solution of the Cauchy problem (2.23) for $u = u^{(k)}$, and χ and Σ are certain continuous functions.

For the OCP (2.23), (2.3), (2.30) with $Q = \mathbb{R}$, $\lambda_u = 0$, and $\lambda_{du} = 0$, consider the problem of improving the process $v^{(k)} = (\psi^{(k)}, u^{(k)}) \in \mathscr{D}$ according to the regularised cost functional $\widetilde{J}(v, v^{(k)})$ (4.10), (4.11). Based on [40] and [66], we formulate the following iterative process, where $v^{(k)} = (\psi^{(k)}, u^{(k)})$ and $v^{(k+1)} = (\psi^{(k+1)}, u^{(k+1)})$ are the input and output admissible processes, respectively, at the kth iteration of the method.

1. Compute the matrix function $\Sigma^{(k)}(t)$ according to the formula (3.21) for some values of $\alpha, \beta < 0$ and $\gamma > 0$, and find the solution $\chi^{(k)}$ of the Cauchy

problem

$$\frac{d\chi^{(k)}(t)}{dt} = -\frac{i}{\hbar}\big(K + V[u^{(k)}(t)] + 2\kappa|\psi^{(k)}(t)|^2\big)\chi^{(k)}(t) + i\kappa\big(\psi^{(k)}(t)\big)^2\chi^{*(k)}(t), \tag{6.3}$$

$$\chi^{(k)}(T) = -\frac{\partial \mathscr{F}}{\partial \psi^*(T)}(\psi^{(k)}(T)). \tag{6.4}$$

2. Find the control $u^{(k+1)}$ by the formula

$$u^{(k+1)}(t) = u^{(k)}(t) + \frac{S(t)}{\gamma_u \hbar}\operatorname{Im}\left[\left\langle \chi^{(k)}(t), \frac{\partial V}{\partial u}\Big|_{u^{(k+1)}(t)} \psi^{(k+1)}(t)\right\rangle + \frac{1}{2}\left\langle \psi^{(k+1)}(t) - \psi^{(k)}(t), \Sigma^{(k)}(t)\frac{\partial V}{\partial u}\Big|_{u^{(k+1)}(t)} \psi^{(k+1)}(t)\right\rangle\right], \tag{6.5}$$

where the function $\psi^{(k+1)}$ is the solution of the Cauchy problem (2.23) with control $u = u^{(k+1)}$.

Theorem 6.1. *For the OCP* (2.23), (2.3), (2.30) *with* $Q = \mathbb{R}$, $\lambda_u = 0$, *and* $\lambda_{du} = 0$ *the method* (6.2)–(6.5) *using the regularisation* (4.10), (4.11) *with* $\gamma_u > 0$ *gives* $J(v^{(k+1)}) \leqslant J(v^{(k)})$.

Remark 6.1. The formulae (2.24), (2.26)–(2.28) describe essentially different potentials in the Gross–Pitaevskii equation. The potential (2.24) is linear in u. Unlike in (2.24), the control in the potential (2.28) enters in polynomial form. The formula (6.5) is obtained from the condition

$$\frac{\partial}{\partial u} H\left(t, \chi^{(k)}(t) + \frac{1}{2}\Sigma^{(k)}(t)\psi^{(k+1)}(t), \psi^{(k+1)}(t), u\right) = 0,$$

where H is defined by (6.1). If V depends linearly on u, then the right-hand side of (6.5) does not contain $u^{(k+1)}(t)$, that is, $u^{(k+1)}$ is easily computed using the formula (6.5). If V depends non-linearly on $u^{(k+1)}$, then the complexity of (6.5) is completely different from that in the linear case.

In [66] it was noted that the function $\Sigma^{(k)}$ can either be computed as the solution of a special Cauchy problem (by analogy with [58]) or be specified in accordance with (3.23). Computations involving the Krotov method with the regularisation (4.10), (4.11) were carried out. In [40] the function $\Sigma^{(k)}$ is determined by (3.23). In [97], p. 34, (Jäger, 2015) it is noted that the Krotov method with linear-quadratic $\varphi^{(k)}$ depends principally on the values of α, β, and γ in (3.23). As discussed in [66], p. 5, one can start from $\Sigma^{(0)} \equiv 0$, decreasing α and β subsequently and increasing γ. If (6.5) does not provide an improvement of the process $v^{(k)}$, then we have to adjust α, β, and γ and repeat the computations.

In [40] and other publications, the regularisation (4.10), (4.11) with parameter γ_u is an important tool. In [97], p. 86, this parameter is called the 'step' in the context of the Krotov method.

For (2.25), the transversality condition (6.4) has the form

$$\chi^{(k)}(T) = -\operatorname{Re}\psi^{(k)}(T) + \frac{1}{2}|\psi^{(k)}(T)|\langle\cos\theta^{(k)}(T)\rangle\left(\frac{\psi^{(k)}(T)}{\psi^{*(k)}(T)} + 3\right)$$

(see [66]). The transversality condition for the terminant $\mathscr{F}_{\psi_{\text{target}}}(\psi(T))$ (2.5) is

$$\chi^{(k)}(T) = \langle\psi_{\text{target}}, \psi^{(k)}(T)\rangle\psi_{\text{target}}. \tag{6.6}$$

In [40] the terminant (2.5) was considered, and the conjugate system was given for the function $p(t) = i\chi(t)$. The condition (6.6) has the form

$$p^{(k)}(T) = i\langle\psi_{\text{target}}, \psi^{(k)}(T)\rangle\psi_{\text{target}}.$$

For OCPs where V depends on u non-linearly, the following simplifications for (6.5) were proposed in [40]:

(a) consider $\Sigma^{(k)} \equiv 0$ (in [40], p. 8, there is a description of how to solve the equation derived from (6.5) for such a variant of the Krotov method);

(b) use $(\partial V/\partial u)\big|_{u^{(k)}(t)}$ instead of $(\partial V/\partial u)\big|_{u^{(k+1)}(t)}$.

Taking both simplifications into account, instead of (6.5) consider the formula

$$u^{(k+1)}(t) \approx u^{(k)}(t) + \frac{S(t)}{\gamma_u\hbar}\operatorname{Im}\left\langle\chi^{(k)}(t), \frac{\partial V}{\partial u}\bigg|_{u^{(k)}(t)}\psi^{(k+1)}(t)\right\rangle. \tag{6.7}$$

This formula was then applied, with the remark that for sufficiently small values of the parameter $\kappa > 0$ in the Gross–Pitaevskii equation the control u varies moderately from one iteration to another. Judging from the computational results described in [40], the Krotov method in the simplified version with (6.7) was successful. The non-linearity of V in u complicates the application of the method to the control of Bose–Einstein condensates.

6.2. Krotov and GRAPE methods in numerical experiments for controlling Bose–Einstein condensates. We briefly explain the GRAPE method [39] (Khaneja, Reiss, Kehlet, Schulte-Herbrüggen, and Glaser, 2005), which is often used to solve OCPs for quantum systems. In this method:

1) the control $u\colon [0,T] \mapsto \mathbb{R}$ is represented as a piecewise constant function

$$u(t) = c_j, \qquad t \in [t_j, t_{j+1}), \quad t_j = j\frac{T}{N}, \quad j = 0, \dots, N, \tag{6.8}$$

where $c_j \in \mathbb{R}$, the control is $c = [c_0, \dots, c_N]$, and $\Delta t = T/N$ is the discretisation step;

2) the OCP is reformulated as the problem of minimising some function $\widetilde{\mathscr{F}}(c)$.

Then gradient optimisation methods can be used. GRAPE is also used [158], [40] in combination with the BFGS (Broyden–Fletcher–Goldfarb–Shanno) method.

For the OCP with linear potential (2.24) and the condition (2.25), the article [66] (Sklarz and Tannor, 2002) shows the results of the second-order Krotov method, which uses (3.23) to compute the function $\Sigma^{(k)}$. As shown in Fig. 3 in [66], the solution was found in almost 30 iterations with successive improvements. Figure 4 in [66] represents the phase $\theta(T)$ relating to the optimised control.

In [40] (Jäger, Reich, Goerz, Koch, and Hohenester, 2014) two OCPs were considered for the Gross–Pitaevskii equation for Bose–Einstein condensates. In the first OCP the goal was to realise a splitting of the Bose–Einstein condensate at time T in such a way that the wavefunction at time T should correspond to the ground state of the two-well potential. The second OCP is for shaking the condensate, where, for the anharmonic single-well potential $V(x-u(t))$, the goal is to move the condensate from the ground state V, which is the state of the system at time $t=0$, to the first excited state. The OCP for splitting a Bose–Einstein condensate was solved independently by:

(a) applying the Krotov method in its simplified version using (6.7);

(b) applying the GRAPE-BFGS method with H_1-regularisation (2.29).

Figure 1 (b, c) in [40] presents the density $n(x,t)=|\psi(x,t)|^2$ in the 'space–time' plane, and shows that splitting of the Bose–Einstein condensate into two parts is achieved. Figure 2 in [40] shows that the Krotov method using (6.7) gives successive improvements whose effectiveness depends on the parameter κ in the Gross–Pitaevskii equation. For $\kappa=\pi/2$ the Krotov method using (6.7) is faster than GRAPE-BFGS-H_1 in terms of the number of Cauchy problems solved: with use of the Krotov method a value of the terminant $\mathscr{F}<10^{-4}$ was obtained, and the complexity equates to solving 100 Cauchy problems. As seen in Fig. 4 in [40], for the OCP with shaking of the Bose–Einstein condensate, the situation is as follows:

(a) solving this OCP turned out to be significantly more difficult (hundreds of Cauchy problems) than solving the OCP for splitting the condensate;

(b) the Krotov method using (6.7) for the problem of shaking a Bose–Einstein condensate yields consistent improvements, and for $\kappa=2\pi$ it shows much better efficiency than GRAPE-BFGS-H_1.

Thus, the first-order Krotov method using (6.7) in the above OCP can be successful for some values of the parameters κ and γ_u.

7. Conclusions

Starting from its development in the late 1970s, quantum control has become a large interdisciplinary area of great importance for science and technology now and in the future. Quantum control exploits methods from diverse areas of mathematics (for example, differential equations, optimal control, functional analysis, group theory, differential geometry, finite-dimensional optimisation, algebra), and has multiple applications to physics and chemistry (control of molecular dynamics, nuclear magnetic resonance, laser chemistry, and so on), quantum computing, and information theory [3]–[23].

The formulation of a control problem for a quantum system involves determining a suitable mathematical model of the controlled system and a cost functional subject to maximisation or minimisation. A mathematical model should effectively describe the controlled dynamics of a physical quantum system. The modelling should involve description of the space of quantum states of the system and their controlled evolution equation, which can be the Schrödinger equation or the Liouville–von Neumann equation for a closed quantum system with a dynamics that is linear with respect to the quantum state, or the non-linear Gross–Pitaevskii equation for

the wavefunction of a Bose–Einstein condensate, a master equation with a control for an open quantum system.

The cost functional to be minimised includes a terminant $\mathscr{F}$ which represents the control goal, along with additional integral terms used to set constraints on the control or on the admissible states, or to improve the quality of the optimisation methods. The terminant for the problem of maximising the mean target observable O of the quantum system at a finite moment of time T has the form

$$\mathscr{F} = -\langle \psi(T), O\psi(T) \rangle,$$

the terminant for the problem of minimising the distance to the target state has the form

$$\mathscr{F} = \|\psi(T) - \psi_{\text{target}}\|^2,$$

and so on. The cost functional can include integral terms

$$\lambda_u \int_0^T \frac{\|u(t)\|^2}{S(t)}\, dt \quad \text{and} \quad \lambda_\psi \int_0^T \langle \psi(t), D(t)\psi(t) \rangle\, dt$$

to restrict the control and the quantum states, respectively. Along with the integral constraint on u, a pointwise constraint $u(t) \in Q \subset \mathbb{R}^m$ can be used. Moreover, one can consider the (in)equality

$$\int_0^T \|u(t)\|^2\, dt\ \{=, \leqslant\}\ E.$$

The final moment of time T is either fixed or free.

Success in finding a quantum optimal control depends on both the skill with which the optimisation model is formulated and the skill with which the optimisation methods are applied. For solving OCPs for quantum systems the following optimisation methods are used:

(a) methods which operate in the function space of the control, in particular, methods based on the Pontryagin maximum principle and the Krotov method;
(b) methods based on reducing an OCP to a finite-dimensional optimisation problem via some parametrisation of the control (for example, GRAPE uses piecewise constant functions, and CRAB uses trigonometric functions);
(c) hybrid methods (for example, a combination of reduction to a finite-dimensional optimisation problem and the Krotov method).

The efficiency of the solution of an OCP depends, in particular, on the quality of the numerical solution of ODEs and PDEs.

The Krotov method has been widely used for quantum control. This method has been applied to the control of molecular dynamics, the realisation of quantum gates, the control of Bose–Einstein condensates, the control of nuclear magnetic resonance, and so on.

The efficiency of the Krotov method depends on the specifics of an OCP, on how the improving function $\varphi^{(k)}$ and the regularisers are defined, and also on the adjustments of the method's parameters. For example, if the potential V depends non-linearly on $u(t)$ in the Gross–Pitaevskii equation (see Section 6), then use of

the Krotov method is more difficult than in the linear case. The function $\varphi^{(k)}$ can be linear or linear-quadratic with respect to the quantum state. If the function $\Sigma^{(k)}$ is defined using (3.23), then the parameters $\alpha, \beta < 0$ and $\gamma > 0$ must be adjusted for improvement of the control.

The Krotov method is one of the *non-local improvement methods*, which, in contrast to *local improvement* methods (for example, steepest descent method, conditional gradient method, and so on), are not limited to small control variations and do not contain a costly procedure for finding the best parametrised variation. In [64] and [65] (Tannor, Kazakov, Orlov, and Somlói, early 1990s), the Krotov method with a linear function $\varphi^{(k)}$ was applied to the OCP (2.1)–(2.4) with $\lambda_\psi = 0$, and it was shown that this gives macro-steps for control improvements, and also that the Krotov method can be faster than the steepest-descent method. The Ho–Rabitz TBQCP method [49] can be faster than the first-order Krotov method.

In a number of publications, including [66], [89], and [91], the Krotov method was used with a linear-quadratic function $\varphi^{(k)}$ with $\Sigma^{(k)}$ defined by (3.23). At the same time, in [58] (Krotov and Feldman, 1983) the function $\Sigma^{(k)}$ was defined as the solution of some special Cauchy problem (3.28), (3.29). In [66] (Sklarz and Tannor), both methods for determining $\Sigma^{(k)}$ were mentioned, although their computations used the method involving (3.23). As was noted in [61], for an OCP outside of quantum control the method with (3.28), (3.29) is more cost expensive than the version with (3.23), but the latter variant can be less effective. Thus, for quantum OCPs a comparison of the efficiency of the two methods is important.

Closed quantum systems describe real quantum systems only approximately, because in practice there is often an unavoidable influence of the external environment. The Krotov method has also been successfully applied to OCPs for open quantum systems ([22], [62], [70], [79], [92]–[96], [104]). This area of research is of great practical interest and must be investigated separately elsewhere.

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