# Optimal and robust quantum control in low dimensional systems 

Kaipeng Liu

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Par<br>LIU Kaipeng

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## Université Bourgogne Franche-Comté

## THESIS

presented by
Kaipeng LIU
to obtain the Degree of
DOCTOR of PHYSICS

# Optimal and robust quantum control in low dimensional systems 

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Date of defense: December 17, 2020

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## Abstract

Optimal control theory (OCT) is the basic and comprehensive method to obtain the optimal solutions of quantum systems controlled by external fields. It provides a powerful set of tools and concepts. One of the goals of the thesis is to design the technique of OCT in two- and three-state quantum systems taking into account losses and robustness, which is of primary importance for the implementation of control techniques in a broad class of platforms.

Based on inverse-engineering techniques and the Pontryagin maximum principle (PMP), we establish and test the different optimal strategies showing how to control the transfer in three-level quantum systems considering energy- and time-minimum optimal solutions taking into account losses. These results, in particular, show that the usual adiabatic passage in such systems, known as stimulated Raman adiabatic passage (STIRAP), which leads to imperfect transfer, can be made exact thus achieving stimulated Raman exact passage (STIREP) while reducing the energy and the duration costs respectively of the controls.

We next combine robustness with optimization. Instead of using a direct optimization procedure from OCT, we develop a technique of geometric optimization that allows the derivation of optimal and robust solutions from an inverse optimization. The method named robust inverse optimization (RIO) allows one to obtain numerical trajectories that can be made as accurate as required. The method is versatile and can be applied to various types of errors and of quantum control problems.

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## Contents

1 Introduction ..... 1
2 From adiabatic passage to exact passage: inverse engineering tech- niques ..... 5
2.1 Definition and properties ..... 6
2.2 Determination of the invariant ..... 9
2.2.1 Generalities ..... 9
2.2.2 Two-state system ..... 10
2.2.3 Three-state system ..... 11
2.3 Single-mode driving: From adiabatic to exact passage ..... 13
2.4 Multi-mode driving ..... 14
2.5 Disscussion ..... 19
3 Techniques of optimal control in quantum systems ..... 21
3.1 Introduction ..... 22
3.2 Lagrange multiplier method ..... 23
3.2.1 Description of the method ..... 23
3.2.2 Example ..... 25
3.2.3 Extension of the method for a time-dependent constraint ..... 27
3.3 Euler-Lagrange principle ..... 28
3.4 The Pontryagin maximum principle ..... 31
3.4.1 The Lagrangian problem with constraints and controls ..... 32
3.4.2 The PMP for the Mayer problem ..... 33
3.4.3 The PMP for the Mayer-Lagrange or Lagrange problems ..... 34
3.5 Optimal control with constrained controls ..... 36
3.6 Maximum Principle and shooting method for the three-level problem ..... 38
3.6.1 Original coordinates ..... 38
3.6.2 Spherical coordinates ..... 42
3.7 Optimal Control by gradient method ..... 48
3.8 Trigonometric expansion for finite-time stimulated Raman exact passage (STIREP) by gradient method ..... 56
3.8.1 Counterintuitive sequence as a starting point in a 8-parameter space ..... 56
3.8.2 Intuitive sequence ..... 61
3.8.3 Gradient method ..... 65
4 Optimal control for dissipative STIREP ..... 69
4.1 Definition of the dissipative driven stimulated Raman system ..... 70
4.2 Optimal control with respect to the energy of the controls constrained by a given admissible loss: energy-optimal dissipative STIREP ..... 72
4.2.1 Construction of the pseudo-Hamiltonian ..... 72
4.2.2 Construction of the optimal trajectories from the Pontryagin max- imum principle ..... 76
4.2.3 Derivation of the pulses and the dynamics ..... 85
4.2.4 Comparison with standard STIRAP and parallel STIRAP ..... 93
4.2.5 Analogy with a pendulum ..... 97
4.3 Optimal control with respect to time constrained by a given admissible loss: time-optimal dissipative STIREP ..... 99
4.3.1 Construction of the pseudo-Hamiltonian ..... 99
4.3.2 Construction of the optimal trajectories from the Pontryagin max- imum principle ..... 102
4.3.3 Derivation of the pulses and the dynamics ..... 115
4.4 Discussion ..... 126
5 Optimal robust quantum control by inverse optimization: robust in- verse optimization (RIO) method ..... 131
5.1 The model and principles of inverse engineering ..... 132
5.2 The single-shot shaped pulse method ..... 134
5.3 Robust population transfer - Figure of merit ..... 136
5.4 The unconstrained optimization problem ..... 138
5.5 Application of RIO for robustness with respect to field inhomogeneities $(\delta=0)$ ..... 140
5.5.1 Complete population transfer ..... 140
5.5.2 Robust trajectory optimal with respect to pulse energy or time ..... 147
5.5.3 Partial population transfer ..... 150
5.6 Discussion ..... 153
6 Conclusion ..... 157
Bibliography ..... 161

## List of Figures

2.1 Diagram of a three-level $\Lambda$-type system for stimulated Raman processes at two-photon resonance. $\Omega_{p}$ and $\Omega_{s}$ denote the pump and Stokes Rabi frequencies, which connect the initial ground state $|1\rangle$ and the excited state $|2\rangle$, the excited state $|2\rangle$ and the final target state $|3\rangle$, respectively.

2.2 Pump $\Omega_{p}$ (upper frame, solid red line) and Stokes $\Omega_{s}$ (upper frame, solid
blue line) (in units of $1 / T)$ and the resulting populations with $\psi(t)$ the
state solution (lower frame) for $k=1$.

2.3 Pump $\Omega_{p}$ (upper frame, solid red line) and Stokes $\Omega_{s}$ (upper frame, solid
blue line) (in units of $1 / T)$ and the resulting populations with $\psi(t)$ the
state solution (lower frame) for $k=2$. ..... 17

2.4 Time-dependence of the parameters $\varphi$ (upper frame, solid cyan line) and
$\theta$ (lower frame, solid magenta line) (in units of $1 / \mathrm{T}$ ) determined from Eq.
(2.52) for $k=1$

3.1 Decimal logarithm of the deviation $\varepsilon$ for various $\Omega_{0} T$ and $\lambda_{1}(0) T$ for
$\omega=-\pi / 2 T$. ..... 43

3.2 Parameters $\varphi$ and $\theta$ (in units of $1 / T$ ) corresponding to Eq. (3.159) ( $\alpha=$
36) (full lines) and Eq. (3.160) $(\beta=2)$ (dashed lines), respectively, as
function of time. ..... 47

3.3 Control pulses $u_{p}$ and $u_{s}$ (in units of $1 / T$ ) corresponding to Eq. (3.159)
( $\alpha=36$ ) (full lines) and Eq. (3.160) $(\beta=2)$ (dashed lines), respectively,
as function of time. ..... 47

3.4 Schematic representation of a control amplitude $u_{k}(t)$, consisting of $N$
discretization steps of duration $\Delta t=T / N$. During each step $n$, the
control amplitude $u_{k, n}$ is constant. The vertical arrows represent the
gradients of $J$, indicating how each amplitude $u_{k, n}$ should be modified in
the next iteration to improve the cost $J$. ..... 50
3.5 Curve of the minimum energy with the variation of $\phi_{3}$ for a given value of $A=0.0879 T$ obtained from multi-mode driving.59
3.6 Pump $u_{p}$ (solid blue line) and Stokes $u_{s}$ (solid red line) pulses with the parameters $\phi_{1}=0.3964, \phi_{3}=-0.1850, \vartheta_{1}=-0.9560$ and $\vartheta_{3}=0.2220$, that gives the smallest energy for a given $A=0.0879 T$ in the chosen 8-parameter restricted space, compared to the pump (dashed cyan line) and Stokes (dashed magenta line) pulses derived by linear multi-mode driving. ..... 60
3.7 Dynamics of the populations resulting from pulses (solid lines) of Figure 3.6. The transfer from state $|1\rangle$ to state $|3\rangle$ is exact due to the chosen boundaries (STIREP) ..... 61
3.8 Curve of the minimum energy with the variation of $\phi_{3}$ for a given $A=$ 0.3750T ensuring the minimum energy in the 6 -parameter space. ..... 62
3.9 Pump $u_{p}$ (solid blue line) and Stokes $u_{s}$ (solid red line) pulses with the parameters $\phi_{1}=0.9874, \phi_{3}=-0.0485, \vartheta_{1}=-0.9030, \vartheta_{3}=0.1330$ realizing the minimum energy in the 6-parameter space, compared to the pump (dashed cyan line) and Stokes (dashed magenta line) pulses derived from (3.155) ..... 63
3.10 Populations resulting from pulses (solid lines) of Figure 3.9 ..... 64
3.11 Dependence of the minimum energy on the time area of the transient population in the excited state. ..... 64
3.12 (a) Energy optimization using the gradient method from initial conditions close to the known minimum energy (with the parameters $\phi_{1}=1, \phi_{3}=$ $0.2, \vartheta_{1}=-0.8, \vartheta_{3}=0.1, \varepsilon=-0.00001$ ). (b) Pump $u_{p}$ (solid blue line) and Stokes $u_{s}$ (solid red line) pulses obtained from the optimization by the gradient method (at the $101^{\text {th }}$ iteration). ..... 67
3.13 (a) Energy optimization using the gradient method from initial conditions very close to the known minimum energy (with the parameters $\phi_{1}=1$, $\phi_{3}=0.2, \vartheta_{1}=-0.9030, \vartheta_{3}=0.1330, \varepsilon=-0.00001$ ). (b) Pump $u_{p}$ (solid blue line) and Stokes $u_{s}$ (solid red line) pulses obtained from the optimization by the gradient method (at the $67^{\text {th }}$ iteration). ..... 68
4.1 Dependence of the optimal energy on the time area of the transient pop- ulation in the excited state. ..... 86
4.2 Time-dependence of the parameters $\varphi$ (upper frame, solid cyan line) and $\theta$ (upper frame, solid magenta line) (in units of $1 / T$ ), the control pulses $u_{p}$ (middle frame, solid red line) and $u_{s}$ (middle frame, solid blue line) (in units of $1 / T$ ) and populations (lower frame) for the time area of the population in the excited state $A=3 T / 8$ corresponding to unconstrained optimal pulses. ..... 87
4.3 Same as Figure 4.2 but for $A=0.2 T$ ..... 88
4.4 Same as Figure 4.2, but for $A=0.07 T$. ..... 89
4.5 Controls for various values of $A=3 T / 8, A=0.3 T$ and $A=0.25 T$. ..... 90
4.6 Same as Figure 4.5 but for $A=0.2 T, A=0.15 T$ and $A=0.1 T$. ..... 90
4.7 Controls for $A=0.08 T$. The pump and Stokes controls appear as over- lapping pulses at the scale of the figure for $A \lesssim 0.08 T$. ..... 91
4.8 Controls for various values of $A<0.08 T$. ..... 91
4.9 Comparison of the dynamics between (a) the energy-optimal STIREP and (b) the standard STIRAP featuring the same time area in the excited state $A \simeq 0.036 T$. ..... 94
4.10 Time-dependence of dynamics for $\Omega_{0}=14.453 / T$. Upper frame: The control pulses $u_{P}(t)$ and $u_{S}(t)$ (in units of $1 / T$ ) according to (4.114). Middle frame: Populations with $\psi(t)$ the state solution. Lower frame: The eigenvalues given by (4.108) (in units of $1 / T$ ). ..... 96
4.11 Contour plots of solution(s) of $\varphi_{0}$ from Eq.(4.153) as a function of $\tilde{\mu}$ and $\tilde{\lambda}_{\theta}$. Absence of solutions is indicated in black. ..... 108
4.12 Contour plots of logarithm to the base 10 of the absolute values of the difference between $\varphi(\pi / 4)$ (from Eq. (4.151b)) and $\varphi_{0}$ (from Eq. (4.153)), as a function of $\tilde{\mu}$ and $\tilde{\lambda}_{\theta}$ for (a) $\tilde{\mu} \leq-8$; and (b) $\left.\left.\tilde{\mu} \in\right]-8,1\right]$. ..... 109
4.13 Dependence of the parameter $\tilde{\lambda}_{\theta}$ on parameter $\tilde{\mu}$ obtained from Table 4.3. 112
4.14 Dependence of the optimal control time $T$ on the area $A$ obtained from Table 4.4. ..... 114
4.15 Time-dependence of the parameters $\varphi$ (upper frame, solid cyan line) and $\theta$ (upper frame, solid magenta line), the control pulses $u_{p}$ (upper middle frame, solid red line) and $u_{s}$ (upper middle frame, solid blue line) (in units of $u_{0}$ ), the projection (in absolute value squared) of the dynam- ics onto the dark state defined with the actual $\theta$ (lower middle frame), and populations (lower frame) for $\tilde{\mu}=0$ and the resulting optimal time $T=2.72 / u_{0}$ shown in Table 4.4 corresponding to unconstrained optimal pulses. ..... 117
4.16 Same as Figure 4.15 , but for $\tilde{\mu}=-5$. ..... 118
4.17 Same as Figure 4.15, but for $\tilde{\mu}=-10.5$. ..... 119
4.18 Same as Figure 4.15, but for $\tilde{\mu}=-20.5$. ..... 120
4.19 Same as Figure 4.15, but for $\tilde{\mu}=-24$. ..... 121
4.20 Comparison of the controls (in units of $u_{0}$ ) for various couples of ( $\left.\tilde{\mu}, \tilde{\lambda}_{\theta}\right)$ and the corresponding optimal time $T$ from Table 4.4. ..... 122
4.21 Same as Figure 4.20 but for larger negative values of $\tilde{\mu}$ and the corresop- nding $\tilde{\lambda}_{\theta}$ and optimal time $T$ from Table 4.4. ..... 122
4.22 Controls (in units of $u_{0}$ ) for $\tilde{\mu}=-11, \tilde{\lambda}_{\theta}=6.2826117$ and optimal time $T=5.06 / u_{0}$ from Table 4.4. ..... 123
4.23 Controls (in units of $u_{0}$ ) for $\tilde{\mu}=-15, \tilde{\lambda}_{\theta}=7.460490528$ and optimal time $T=5.96 / u_{0}$ from Table 4.4. ..... 123
4.24 Controls (in units of $u_{0}$ ) for $\tilde{\mu}=-18, \tilde{\lambda}_{\theta}=8.22979283925$ and optimal time $T=6.57 / u_{0}$ from Table 4.4. ..... 124
4.25 Controls (in units of $u_{0}$ ) for $\tilde{\mu}=-19, \tilde{\lambda}_{\theta}=8.47035156274$ and optimal time $T=6.74 / u_{0}$ from Table 4.4. ..... 124
4.26 Controls (in units of $u_{0}$ ) for $\tilde{\mu}=-22, \tilde{\lambda}_{\theta}=9.1535580319427$ and optimal time $T=7.28 / u_{0}$ from Table 4.4. ..... 125
4.27 Controls (in units of $u_{0}$ ) for $\tilde{\mu}=-24, \tilde{\lambda}_{\theta}=9.581657691415900$ and optimal time $T=7.62 / u_{0}$ from Table 4.4. ..... 125
4.28 Pulse amplitudes for the three examples (i), (ii), (iii) described in the text. For (ii) and (iii), we have only plotted $u_{0}$. ..... 126
4.29 Final population transfer as a function of the deviation $\varepsilon$ for energy- optimal dissipative STIREP with respect to various time area of the transient population in the excited state $A=0.375 T$ (unconstrained case) (solid blue line), $A=0.1 T$ (solid red line) and $A=0.05 T$ (dashed green line), respectively ..... 127
4.30 Comparison of final population transfer as a function of the deviation $\varepsilon$ between time-optimal dissipative STIREP (solid lines) and STIRAP (dashed lines) with respect to various time area of the transient popula- tion in the excited state $A=0.375 T$ (unconstrained case) (blue lines), $A=0.1 T$ (red lines) and $A=0.05 T$ (green lines), respectively. ..... 128
4.31 Stokes $u_{s}$ (upper frame, solid red line) and Pump $u_{p}$ (upper frame, solid black line) (in units of $1 / T$ ) for STIRAP, the projection (in absolute value squared) of the dynamics onto the dark state defined with the actual $\theta$ (middle frame) and the resulting populations with $\psi(t)$ the state solution (lower frame) in a situation of time area of the transient population in the excited state $A=0.375 T$ ..... 129
4.32 Same as Figure 4.31 but for $A=0.1 T$. ..... 130
4.33 Same as Figure 4.31 but for $A=0.05 T$. ..... 130
5.1 Symbolic path diagrams giving the construction of the $O_{n}$ integrals. Thesymbol $e$ stands for $e(t), e^{\prime}$ for $e\left(t^{\prime}\right)$, and so on. For instance, for $n=3$,the diagram features four paths. Its extension for larger $n$ is direct.135

# 5.2 Optimal robust trajectory $\widetilde{\theta}(\gamma)$ corresponding to $\lambda_{1} \approx-1.1150$ and $\lambda_{2} \approx$ -0.3047 (leading to $\gamma_{f}=5 \pi / 3$ ). 

5.3 Time-dependence of Rabi frequency $\Omega$ (upper frame, solid blue line) and
detuning $\Delta$ (upper frame, solid red line) and the resulting populations
$P_{j}, j=1,2$ (lower frame) for complete population transfer for the time
parameterization $\gamma(t)$ (5.74). ..... 147
5.4 Resulting detuning and dynamics of the populations $P_{j}, j=1,2$, for
robust time-optimal control [obtained for a flat pulse of Rabi frequency
$\Omega_{0}$ according to (5.85)] showing the complete population transfer with
the optimal time $T_{\min } \approx 5.84 / \Omega_{0}$. ..... 150
5.5 Optimal robust trajectory $\widetilde{\theta}(\gamma)$ corresponding to $\lambda_{1} \approx-1.6974$ and $\lambda_{2} \approx$ -0.6465 (leading to $\gamma_{f} \approx 1.48 \pi$ ). ..... 152
5.6 Time-dependence of Rabi frequency $\Omega$ (upper frame, solid blue line) and detuning $\Delta$ (upper frame, solid red line) and the resulting populations $P_{j}, j=1,2$ (lower frame) for partial population transfer targeting the half superposition state (up to a global phase). ..... 152
5.7 Detuning and dynamics of the populations $P_{j}, j=1,2$, for robust time- optimal control (for a flat pulse of Rabi frequency $\Omega_{0}$ ) showing the half superposition. We obtain the optimal time $T_{\min } \approx 4.05 / \Omega_{0}$. ..... 153
5.8 Comparison of the robustness profile given by the RIO trajectories (solid cyan lines) with $\pi / 2$-pulse (solid magenta lines). Upper frame: Transition probability as a function of $\alpha$. Lower frame: Logarithmic scale of the deviation targeting at half superposition state. ..... 154

## List of Tables

4.1 Losses of the model (4.1) (4.2a) and the approximate model (4.4) (4.2b) for different and relatively low ratios $\Gamma / u_{\max }$ (obtained for example from standard STIRAP with Gaussian pulses with the peak of the pulses set to $\left.u_{\max }=6 / T\right)$. The approximate model provides reliable losses for the ratio $\Gamma / u_{\max }$ considered.71
4.2 Optimal values of $m, \varphi_{0}$ and the corresponding energy $\mathcal{E}$ for various $A$. The value $A=0.0879 T$ corresponds to the multi-mode driving for $k=1$ (see Section 2.4). The peak value $u_{\max }$ and the pulse area $\mathcal{A}=$ $\int_{0}^{T} d s \sqrt{u_{p}^{2}+u_{s}^{2}}$ are also given. The pulse area for the Rabi frequency is $2 \mathcal{A}$.

4.3 Values of couple $\left(\tilde{\mu}, \tilde{\lambda}_{\theta}\right)$ satisfying $\left|\varphi(\pi / 4)-\varphi_{0}\right|=0$, and the correspond
ing values of $\log _{10}\left|\varphi(\pi / 4)-\varphi_{0}\right|$. ..... 111
4.4 Resulting values of $\varphi_{0}$ (i.e. $\varphi(T / 2)$ ), the time area of the transient population in the excited state $A$ and the optimal control time $T$ for various couples of $\left(\tilde{\mu}, \tilde{\lambda}_{\theta}\right)$. The pulse energy $\mathcal{E}$, Eq. (4.7), corresponds to the value given for optimal time but in units of $u_{0}$. The pulse area $\mathcal{A}=\int_{0}^{T} d s \sqrt{u_{p}^{2}+u_{s}^{2}}$ is also given (which corresponds in fact to the value of $u_{0} T$ ).

## CHAPTER 1

## Introduction

With the continuous deepening of research on emergent quantum information processing, the manipulation of quantum systems has shown an increasing significance in quantum computing, quantum metrology, and high-resolution spectroscopy [1, 2, 3], and has brought tremendous changes and broad application prospects to the fields from laser spectroscopy, molecular and solid-state physics to nuclear magnetic resonance (NMR) and magnetic resonance imaging $[4,5,6,7,8]$. Thus achieving the manipulation of quantum systems, i.e., finding some achievable control fields to drive the controlled system evolving from the given initial state to the desired target state, plays an important role in modern science and technology. Laser pulses are one of the most effective methods to control the dynamics of quantum systems, and many techniques have been developed for such investigations, like resonant pulses, rapid adiabatic passage (RAP), stimulated Raman adiabatic passage (STIRAP) and the corresponding variants $[9,10,11,12,13,14,15]$.

A well-known problem is to design solutions of quantum systems driven by external laser pulses that are optimal with respect to practical costs such as energy and duration [16]. The necessary conditions of optimality were established by Pontryagin via a maximum principle [17]. Based on this approach, various optimal quantum problems have been solved from low- $[18,19,20,21]$ to large-dimensional [22, 8] systems. In a general sense, OCT is a optimal control method providing a direct optimization procedure, and is being widely valued by scientific researchers [23, 24, 25, 26, 27]. This corresponds to finding the optimal strategy, which allows the system to reach a specified target state while minimizing a given cost. There are two central steps in a typical workflow, first solving equations of motion resulting from the necessary condition of the PMP, and next selecting the optimal one(s) minimizing the cost. However, the practical application requires finding a simple and manageable representation of the problem (i.e. the relevant coordinates), which does not often yield closed-form formulas. Detailed calculation and examples will be presented in Chapters 3 and 4.

Solutions that additionally feature robustness have become a major issue in quantum physics, especially in quantum information processing, where ultra high-fidelity solutions
are required (typically with relative errors not above $10^{-4}$ ) [28]. Small imperfections in the design can cause fatal deviations of the performance. Robustness can be specifically taken into account using adiabatic [11, 12], composite [29, 30, 31], combined [32] or shortcut to adiabaticity (STA) $[33,34,35,36]$ techniques. However, these methods are not optimal and usually cost non-necessary energy and time. Combining robustness constraints with the optimization methods has thus become a major challenge [5]. OCT offers some solutions. More precisely, gradient method based on time discretization with thousands of parameters to be optimized [37] leads to very different results depending on the algorithm and the initial condition used, and rarely provides a global optimal solution. Alternative techniques involving from a few tens [38] to a few [39] parameters to be optimized have been developed, but they do not provide global optimal solutions in principle since they are based on restricted parametrizations. A recent proposal using Pontryagin's maximum principle in an extended Hilbert space [40] allows an elegant integration of the robustness constraints, but leads to complicated systems to solve, only tractable for very simple targets, typically population transfers. A geometrical approach has been shown to provide optimal single-qubit phase gates [41]. All these methods use a direct optimization procedure, i.e. with the dynamical equations as constraints, which makes complicated the simultaneous integration of the robustness constraints. We next propose an alternative method of geometric optimization based on inverse engineering, mainly applying the single-shot shaped pulse method (SSSP) [35] for incorporating robustness, which we name robust inverse optimization (RIO). Details are presented in Chapter 5.

The present thesis is organized as follows:
In Chapter 2 we present the invariant-based inverse engineering techniques containing basic concepts, practical applications in two-/three-state systems and interpretations of exact passages from single-/multi-mode driving. We highlight that inverse engineering techniques accelerate the process while simultaneously leading to an exact transfer via a trajectory determined by a choice of the time dependence of the parameters satisfying the appropriate boundaries. Applied to control in $\Lambda$ systems, it is referred to as stimulated Raman exact passage (STIREP) in contrast to the approximate adiabatic passage of STIRAP.

In Chapter 3 we describe the general techniques of OCT, which allow one to obtain optimal trajectories in the parameter space. A general statement, basic approaches and involved concepts, such as the Lagrangian multiplier method, the Euler-Lagrange principle, the Pontryagin maximum principle (PMP), the shooting method and the gradient method are presented. Moreover, we show how PMP can be used to establish the formulation for low-dimension optimal quantum problems which can be solved analytically.

In Chapter 4 we apply PMP to determine solutions of a $\Lambda$ three-state system driven by pump and Stokes pulses in a Raman configuration, which are optimal with respect to practical costs, including energy and duration, taking loss of the excited state into consideration. We first investigate the energy-minimum optimal control solution under a constraint of a given admissible loss in a fixed duration. We obtain the optimal controls featuring an intuitive pulse sequence for relatively large values of loss and a fully overlapping pulse sequence for small losses characterized by shorter duration and more intense amplitude. We next turn our attention to the problem of minimizing the time, still in such a three-state system, and obtain an intuitive shaping again for large losses. The optimal controls in the case of small losses show a transient counterintuitive pulse sequence resembling to the STIRAP sequence. The difference is that this transient counterintuitive sequence is sandwiched between fast intuitive sequences.

In Chapter 5, we take robustness into account. We develop a technique that allows the derivation of optimal and robust solutions achieving stipulated population transfer of two-state quantum systems. Instead of considering the dynamical equations of the problem as constraints of the optimization (as shown in Chapters 3 and 4), we implement an inverse optimization: we determine in the independent dynamical variable space exact-fidelity trajectories constrained to robust solutions; the control fields are then derived from the obtained robust geodesics and the inverted dynamical equations where the parameters of the control fields are expressed as functions of the dynamical variables (that is the inverse-engineering representation of the Schrödinger equation). This RIO method is applied to design optimal control fields producing robust (complete and half) population transfers with respect to the pulse area. Such method can be applied to other quantum control problems such as quantum gates and other types of errors.

The discussion in Chapter 6 concludes the thesis.

## From adiabatic passage to exact passage: inverse engineering techniques

## Contents

2.1 Definition and properties ..... 6
2.2 Determination of the invariant ..... 9
2.2.1 Generalities ..... 9
2.2.2 Two-state system ..... 10
2.2.3 Three-state system ..... 11
2.3 Single-mode driving: From adiabatic to exact passage ..... 13
2.4 Multi-mode driving ..... 14
2.5 Disscussion ..... 19

One emblematic adiabatic passage technique is the stimulated Raman adiabatic passage (STIRAP), as shown in Figure 2.1. It features a $\Lambda$ quantum system driven by two-photon resonant pump and Stokes pulses, switched on and off in a counterintuitive sequence (first Stokes, next pump pulse, with a sufficient overlap). Such process, as all adiabatic processes, is based on the adiabatic theorem, which requires an infinite time of interaction (adiabatic limit). In practice they operate in a relatively long time in order to preserve a sufficient adiabaticity during the dynamics. As a consequence, adiabatic techniques, while in principle robust to systematic deviations of the parameters [13], are approximate and do not lead to exact transfer in a finite time.

For Raman systems, the upper state usually features dissipation (spontaneous emission) and the STIRAP dynamics, in the adiabatic limit, follows an instantaneous dark eigenstate, which is immune to losses. However, in practice, operating in a finite time, the STIRAP process is lossy since it cannot follow exactly the dark state (see Section 2.3). Inverse engineering and other methods based on adiabatic techniques and composite pulses [12, 42, 43, 44] have been proposed to solve such issues, i.e., to accelerate the
process with an ultra-high fidelity and a robustness with respect to systematic errors. Finding an optimal compromise between fast, exact, immune to loss and robust process is still an open question, which we partially solve in this thesis.

In this chapter, we describe the invariant-based inverse engineering technique. It allows an exact transfer via a trajectory determined by a choice of the time dependence of the parameters satisfying the appropriate boundaries. This is referred to as stimulated Raman exact passage (STIREP) in contrast to the approximate adiabatic passage of STIRAP. Section 2.1 and Section 2.2 present some basic concepts of the invariant, in particular, Subsection 2.2 .2 and Subsection 2.2 .3 show the applications in two- and three-state systems, respectively. Exact passages are interpreted in Sections 2.3 and 2.4 in terms of single-mode and multi-mode generalizing the eigenstate(s) approximately followed during an adiabatic dynamics. Discussion is provided in Section 2.5.


Figure 2.1: Diagram of a three-level $\Lambda$-type system for stimulated Raman processes at two-photon resonance. $\Omega_{p}$ and $\Omega_{s}$ denote the pump and Stokes Rabi frequencies, which connect the initial ground state $|1\rangle$ and the excited state $|2\rangle$, the excited state $|2\rangle$ and the final target state $|3\rangle$, respectively.

### 2.1 Definition and properties

The Lewis-Riesenfeld dynamical invariant extends the concept of classical invariant to quantum mechanics [45]. It is formulated by an Hermitian operator $I(t)$ whose quantum average is constant during the dynamics:

$$
\begin{equation*}
\langle\psi(t)| I(t)|\psi(t)\rangle=\text { const } \tag{2.1}
\end{equation*}
$$

with $|\psi(t)\rangle$ solution of the Schrödinger equation $i \frac{d|\psi\rangle}{d t}=H(t)|\psi(t)\rangle$ (in a system of units such that $\hbar=1$ ) and $H(t)$ the Hamiltonian of the system. Combining these equations leads to

$$
\begin{align*}
\frac{d}{d t}\langle\psi(t)| I(t)|\psi(t)\rangle & =\left(\frac{d}{d t}\langle\psi(t)|\right) I(t)|\psi(t)\rangle+\langle\psi(t)| \frac{\partial I(t)}{\partial t}|\psi(t)\rangle+\langle\psi(t)| I(t)\left(\frac{d}{d t}|\psi(t)\rangle\right) \\
& =0, \tag{2.2}
\end{align*}
$$

i.e., from $\frac{d}{d t}\langle\psi(t) \mid \psi(t)\rangle=\left(\frac{d}{d t}\langle\psi(t)|\right)|\psi(t)\rangle+\langle\psi(t)|\left(\frac{d}{d t}|\psi(t)\rangle\right)=0$ giving $i \frac{d}{d t}\langle\psi(t)|=$ $-\langle\psi(t)| H$, to

$$
\begin{equation*}
i \frac{\partial I}{\partial t}=[H(t), I(t)] . \tag{2.3}
\end{equation*}
$$

The eigenvectors $\left|\phi_{n}(t)\right\rangle$ of the invariant, with $\lambda_{n}$ the eigenvalues,

$$
\begin{equation*}
I(t)\left|\phi_{n}(t)\right\rangle=\lambda_{n}\left|\phi_{n}(t)\right\rangle \tag{2.4}
\end{equation*}
$$

allow one to define a basis of dynamical modes on which we can expand the solution of the Schrödinger equation as

$$
\begin{equation*}
|\psi(t)\rangle=\sum_{n} c_{n, i} e^{i \xi_{n}(t)}\left|\phi_{n}(t)\right\rangle \tag{2.5}
\end{equation*}
$$

with the connection at the initial time $t_{i}$

$$
\begin{equation*}
c_{n, i}=\left\langle\phi_{n}\left(t_{i}\right) \mid \psi\left(t_{i}\right)\right\rangle \tag{2.6}
\end{equation*}
$$

and the Lewis-Riesenfeld phases

$$
\begin{equation*}
\xi_{n}(t)=\int_{t_{i}}^{t} d s\left\langle\phi_{n}(s)\right| i \frac{\partial}{\partial s}-H(s)\left|\phi_{n}(s)\right\rangle . \tag{2.7}
\end{equation*}
$$

The invariant features the following properties:
Its eigenvalues $\lambda_{n}$ are time independent.
We show it by considering the scalar product on (2.3):

$$
\begin{equation*}
i\left\langle\phi_{m}(t)\right| \frac{\partial I}{\partial t}\left|\phi_{n}(t)\right\rangle=\left(\lambda_{n}-\lambda_{m}\right)\left\langle\phi_{m}(t)\right| H(t)\left|\phi_{n}(t)\right\rangle \tag{2.8}
\end{equation*}
$$

and on the time derivative of (2.4):

$$
\begin{equation*}
\left\langle\phi_{m}(t)\right| \frac{\partial I}{\partial t}\left|\phi_{n}(t)\right\rangle=\left(\lambda_{n}-\lambda_{m}\right)\left\langle\phi_{m}(t)\right| \frac{\partial\left|\phi_{n}(t)\right\rangle}{\partial t}+\frac{\partial \lambda_{n}}{\partial t} \delta_{m n} \tag{2.9}
\end{equation*}
$$

Considering $m=n$, or a degenerate eigenvalue, gives from (2.8)

$$
\begin{equation*}
\left\langle\phi_{n}(t)\right| \frac{\partial I}{\partial t}\left|\phi_{n}(t)\right\rangle=0 \tag{2.10}
\end{equation*}
$$

and from (2.9)

$$
\begin{equation*}
\left\langle\phi_{n}(t)\right| \frac{\partial I}{\partial t}\left|\phi_{n}(t)\right\rangle=\frac{\partial \lambda_{n}}{\partial t} \tag{2.11}
\end{equation*}
$$

from which we conclude $\frac{\partial \lambda_{n}}{\partial t}=0$.
For $m \neq n$, one concludes, only if the eigenvalue is non-degenerate:

$$
\begin{equation*}
i\left\langle\phi_{m}(t)\right| \frac{\partial}{\partial t}\left|\phi_{n}(t)\right\rangle=\left\langle\phi_{m}(t)\right| H(t)\left|\phi_{n}(t)\right\rangle \tag{2.12}
\end{equation*}
$$

The eigenvalues $\lambda_{n}$ are real and the eigenvectors $\left|\phi_{n}(t)\right\rangle$ form an orthonormal basis, i.e. $\left\langle\phi_{m}(t) \mid \phi_{n}(t)\right\rangle=\delta_{m n}$ since $I(t)$ is Hermitian.

If $|\psi(t)\rangle$ is solution of the Schrödinger equation $i \frac{\partial|\psi\rangle}{\partial t}=H(t)|\psi(t)\rangle$, then $I(t)|\psi(t)\rangle$ is also solution.

This is shown by calculating:

$$
\begin{align*}
i \frac{\partial}{\partial t} I(t)|\psi(t)\rangle & =i \frac{\partial I}{\partial t}|\psi(t)\rangle+i I(t) \frac{\partial}{\partial t}|\psi(t)\rangle \\
& =[H(t), I(t)]|\psi(t)\rangle+I(t) H|\psi(t)\rangle \\
& =H(t) I(t)|\psi(t)\rangle \tag{2.13}
\end{align*}
$$

To show (2.5)-(2.7), we use the spectral expansion

$$
\begin{equation*}
I(t)=\sum_{n} \lambda_{n}\left|\phi_{n}(t)\right\rangle\left\langle\phi_{n}(t)\right|, \tag{2.14}
\end{equation*}
$$

and calculate, defining $c_{n}(t)=\left\langle\phi_{n}(t) \mid \psi(t)\right\rangle$, and using (2.13):

$$
\begin{aligned}
i \frac{\partial}{\partial t} I(t)|\psi(t)\rangle & =i \frac{\partial}{\partial t} \sum_{n} \lambda_{n}\left|\phi_{n}(t)\right\rangle\left\langle\phi_{n}(t) \mid \psi(t)\right\rangle \\
& =i \sum_{n} \lambda_{n} \frac{\partial}{\partial t} c_{n}(t)\left|\phi_{n}(t)\right\rangle \\
& =i \sum_{n} \lambda_{n}\left[c_{n}(t) \frac{\partial\left|\phi_{n}(t)\right\rangle}{\partial t}+\frac{\partial c_{n}(t)}{\partial t}\left|\phi_{n}(t)\right\rangle\right] \\
& =H(t) I(t)|\psi(t)\rangle \\
& =\sum_{n} \lambda_{n} c_{n}(t) H(t)\left|\phi_{n}(t)\right\rangle
\end{aligned}
$$

Projecting on the bra $\left\langle\phi_{m}(t)\right|$, we obtain:

$$
\begin{aligned}
& i \lambda_{m} c_{m}(t)\left\langle\phi_{m}(t)\right| \frac{\partial\left|\phi_{m}(t)\right\rangle}{\partial t}+\sum_{n \neq m} i \lambda_{n} c_{n}(t)\left\langle\phi_{m}(t)\right| \frac{\partial\left|\phi_{n}(t)\right\rangle}{\partial t}+i \lambda_{m} \frac{\partial c_{m}(t)}{\partial t} \\
= & \sum_{n} \lambda_{n} c_{n}(t)\left\langle\phi_{m}(t)\right| H(t)\left|\phi_{n}(t)\right\rangle
\end{aligned}
$$

i.e. using (2.12),

$$
\frac{\partial c_{m}(t)}{\partial t}=i c_{m}(t)\left\langle\phi_{m}(t)\right|\left[i \frac{\partial}{\partial t}-H(t)\right]\left|\phi_{m}(t)\right\rangle
$$

The solution reads

$$
\begin{equation*}
c_{m}(t)=c_{m, i} e^{i \xi_{m}(t)} \tag{2.15}
\end{equation*}
$$

with the $c_{m, i}$ and the phase $\xi_{m}(t)$ given by (2.6) and (2.7) respectively.
We remark that the density matrix $\rho(t)=|\psi(t)\rangle\langle\psi(t)|$ is itself an invariant since it is solution of the Von Neumman Equation:

$$
\begin{equation*}
i \frac{\partial \rho}{\partial t}=[H(t), \rho(t)] \tag{2.16}
\end{equation*}
$$

### 2.2 Determination of the invariant

### 2.2.1 Generalities

We assume that the Hamiltonian has the general form

$$
\begin{equation*}
H(t)=\sum_{i} \Omega_{i}(t) T_{i} \tag{2.17}
\end{equation*}
$$

where the set $\left\{T_{i}\right\}$ of the $N$ Hermitian and time independent operators $T_{i}^{\dagger}=T_{i}$ generates a close algebra:

$$
\begin{equation*}
\left[T_{i}, T_{j}\right]=\sum_{k} C_{i j}^{k} T_{k}, \quad C_{j i}^{k}=-C_{i j}^{k}, \quad C_{i j}^{k}=C_{j k}^{i}=C_{k i}^{j} \tag{2.18}
\end{equation*}
$$

Since the $T_{i}$ 's are Hermitian, the $C_{i j}^{k}$ 's are purely imaginary.

We seek for an invariant as an element of the algebra:

$$
\begin{equation*}
I(t)=\sum_{i} \alpha_{i}(t) T_{i} \tag{2.19}
\end{equation*}
$$

Since $I(t)$ and the $T_{i}$ 's are Hermitian, the $\alpha_{i}(t)$ 's are real.
We can calculate:

$$
\begin{aligned}
i \frac{\partial}{\partial t} I(t)=i \sum_{i} \dot{\alpha}_{i} T_{i} & =[H(t), I(t)] \\
& =\sum_{i j} \Omega_{i}(t) \alpha_{j}(t)\left[T_{i}, T_{j}\right] \\
& =\sum_{i j k} \Omega_{i}(t) \alpha_{j}(t) C_{i j}^{k} T_{k} \\
& =\sum_{i j k} \Omega_{k}(t) \alpha_{j}(t) C_{k j}^{i} T_{i}
\end{aligned}
$$

We then get the coefficients $\alpha_{i}$ 's solution of the equation:

$$
\begin{equation*}
i \dot{\alpha}_{i}=\sum_{j} \beta_{i j}(t) \alpha_{j}(t), \quad \beta_{i j}(t)=\sum_{k} \Omega_{k}(t) C_{k j}^{i} \tag{2.20}
\end{equation*}
$$

The matrix $B(t)=\left[\beta_{i j}(t)\right]_{i=1 . . N, j=1 . . N}$ is Hermitian (with purely imaginary elements) since $\beta_{j i}(t)=\sum_{k} \Omega_{k}(t) C_{k i}^{j}=-\sum_{k} \Omega_{k}(t) C_{k j}^{i}=-\beta_{i j}(t)$. Denoting

$$
\begin{equation*}
|\alpha(t)\rangle=\left[\alpha_{1}(t), \alpha_{2}(t), \cdots, \alpha_{N}(t)\right]^{\top} \tag{2.21}
\end{equation*}
$$

where the superscript $T$ denotes the transpose of the vector, the equation reads:

$$
\begin{equation*}
i \frac{d}{d t}|\alpha(t)\rangle=B(t)|\alpha(t)\rangle, \quad B^{\dagger}(t)=B(t) \tag{2.22}
\end{equation*}
$$

The propagator of this equation is then unitary, which implies:

$$
\begin{equation*}
\langle\alpha(t) \mid \alpha(t)\rangle=\sum_{i} \alpha_{i}^{2}(t)=\text { const. }=\left\langle\alpha\left(t_{i}\right) \mid \alpha\left(t_{i}\right)\right\rangle \tag{2.23}
\end{equation*}
$$

### 2.2.2 Two-state system

We consider the two-state system:

$$
H=\frac{1}{2}\left[\begin{array}{cc}
-\Delta & \Omega  \tag{2.24}\\
\Omega & \Delta
\end{array}\right]=-\frac{1}{2} \Delta \sigma_{z}+\frac{1}{2} \Omega \sigma_{x}
$$

with $\Delta$ and $\Omega$ real. This corresponds to the $\mathrm{SU}(2)$ Lie algebra $(N=3)$ with

$$
\begin{equation*}
T_{1} \equiv \sigma_{x} / 2, \quad T_{2} \equiv \sigma_{y} / 2, \quad T_{3} \equiv \sigma_{z} / 2, \quad\left[T_{1}, T_{2}\right]=i T_{3} \tag{2.25}
\end{equation*}
$$

(and all cyclic permutations of the indices), with $\Omega_{1}=\Omega, \Omega_{2}=0, \Omega_{3}=\Delta, C_{12}^{3}=i$ and $C_{12}^{k \neq 3}=0$. The matrix $B$ reads

$$
B=\left[\begin{array}{ccc}
0 & -i \Delta & 0  \tag{2.26}\\
i \Delta & 0 & -i \Omega \\
0 & i \Omega & 0
\end{array}\right]
$$

To parametrize the three angles $\alpha_{i}$ 's of the expansion (2.19), we need two (timedependent) angles $\varphi, \theta$ and a constant $\alpha_{0}$ in order to satisfy (2.23), $\alpha_{1}^{2}+\alpha_{2}^{2}+\alpha_{3}^{2}=\alpha_{0}^{2}$ :

$$
\begin{equation*}
\alpha_{1}=\alpha_{0} \cos \varphi \sin \theta, \quad \alpha_{2}=\alpha_{0} \sin \varphi \sin \theta, \quad \alpha_{3}=\alpha_{0} \cos \theta \tag{2.27}
\end{equation*}
$$

Equation (2.22) leads to

$$
\begin{equation*}
\dot{\theta}=-\Omega \sin \varphi, \quad \Delta=\dot{\varphi}-\dot{\theta} \operatorname{cotan} \varphi \operatorname{cotan} \theta \tag{2.28}
\end{equation*}
$$

which connects the angles $(\theta, \varphi)$ to the pulse parameters $(\Omega, \Delta)$. The invariant $I(t)$ reads with the parametrization (2.27):

$$
I(t)=\frac{\alpha_{0}}{2}\left[\begin{array}{cc}
\cos \theta & \sin \theta e^{-i \varphi}  \tag{2.29}\\
\sin \theta e^{i \varphi} & -\cos \theta
\end{array}\right]
$$

giving the dynamical modes and the associated phases

$$
\begin{gather*}
\left|\phi_{1}(t)\right\rangle=\left[\begin{array}{c}
\cos (\theta / 2) e^{-i \varphi / 2} \\
\sin (\theta / 2) e^{i \varphi / 2}
\end{array}\right], \quad \lambda_{1}=\frac{\alpha_{0}}{2}, \quad \dot{\xi}_{1}=-\frac{\Omega}{2} \frac{\cos \varphi}{\sin \theta}=\frac{\dot{\theta}}{2} \frac{\operatorname{cotan} \varphi}{\sin \theta},  \tag{2.30a}\\
\left|\phi_{2}(t)\right\rangle=\left[\begin{array}{c}
-\sin (\theta / 2) e^{-i \varphi / 2} \\
\cos (\theta / 2) e^{i \varphi / 2}
\end{array}\right], \quad \lambda_{2}=-\frac{\alpha_{0}}{2}, \quad \dot{\xi}_{2}=\frac{\Omega}{2} \frac{\cos \varphi}{\sin \theta}=-\frac{\dot{\theta}}{2} \frac{\operatorname{cotan} \varphi}{\sin \theta} . \tag{2.30b}
\end{gather*}
$$

### 2.2.3 Three-state system

We consider the (both one-photon and two-photon) resonant three-state system (in the rotating wave approximation):

$$
H=\frac{\hbar}{2}\left[\begin{array}{ccc}
0 & \Omega_{p}(t) & 0  \tag{2.31}\\
\Omega_{p}(t) & 0 & \Omega_{s}(t) \\
0 & \Omega_{s}(t) & 0
\end{array}\right]=\frac{\hbar}{2}\left(\Omega_{p}(t) K_{1}+\Omega_{s}(t) K_{2}\right)
$$

with $\Omega_{p}$ and $\Omega_{s}$ real, where $K_{1}, K_{2}$, and $K_{3}$ are angular-momentum operators for spin 1 :

$$
K_{1}=\left[\begin{array}{lll}
0 & 1 & 0  \tag{2.32}\\
1 & 0 & 0 \\
0 & 0 & 0
\end{array}\right], \quad K_{2}=\left[\begin{array}{lll}
0 & 0 & 0 \\
0 & 0 & 1 \\
0 & 1 & 0
\end{array}\right], \quad K_{3}=\left[\begin{array}{ccc}
0 & 0 & -i \\
0 & 0 & 0 \\
i & 0 & 0
\end{array}\right]
$$

satisfying the commutation relations

$$
\begin{equation*}
\left[K_{1}, K_{2}\right]=i K_{3}, \quad\left[K_{2}, K_{3}\right]=i K_{1}, \quad\left[K_{3}, K_{1}\right]=i K_{2} \tag{2.33}
\end{equation*}
$$

This corresponds to the $\mathrm{SU}(2)$ Lie algebra $(N=3)$ with

$$
\begin{equation*}
T_{1} \equiv K_{1}, \quad T_{2} \equiv K_{2}, \quad T_{3} \equiv K_{3}, \quad\left[T_{1}, T_{2}\right]=i T_{3} \tag{2.34}
\end{equation*}
$$

(and all cyclic permutations of the indices), with $\Omega_{1}=\Omega_{p}, \Omega_{2}=\Omega_{s}, \Omega_{3}=0, C_{12}^{3}=i$ and $C_{12}^{k \neq 3}=0$. The matrix $B$ reads

$$
B=\frac{\hbar}{2}\left[\begin{array}{ccc}
0 & 0 & i \Omega_{2}  \tag{2.35}\\
0 & 0 & -i \Omega_{1} \\
-i \Omega_{2} & i \Omega_{1} & 0
\end{array}\right]
$$

To parametrize the three angles $\alpha_{i}$ 's of the expansion (2.19), we need again two (timedependent) angles $\varphi, \theta$ and a constant $\alpha_{0}$ in order to satisfy (2.23), $\alpha_{1}^{2}+\alpha_{2}^{2}+\alpha_{3}^{2}=\alpha_{0}^{2}$ :

$$
\begin{align*}
& \alpha_{1}=\alpha_{0} \sin \theta \cos \varphi  \tag{2.36a}\\
& \alpha_{2}=\alpha_{0} \cos \theta \cos \varphi  \tag{2.36b}\\
& \alpha_{3}=\alpha_{0} \sin \varphi \tag{2.36c}
\end{align*}
$$

Equation (2.22) allows one to write the Rabi frequencies as functions of angles $(\theta, \varphi)$ :

$$
\begin{align*}
& \Omega_{1} / 2 \equiv \Omega_{p} / 2=\dot{\varphi} \cos \theta+\dot{\theta} \sin \theta \operatorname{cotan} \varphi  \tag{2.37a}\\
& \Omega_{2} / 2 \equiv \Omega_{s} / 2=-\dot{\varphi} \sin \theta+\dot{\theta} \cos \theta \operatorname{cotan} \varphi \tag{2.37b}
\end{align*}
$$

The invariant $I(t)$ reads with the parametrization (2.36a):

$$
I(t)=\alpha_{0}\left[\begin{array}{ccc}
0 & \sin \theta \cos \varphi & -i \sin \varphi  \tag{2.38}\\
\sin \theta \cos \varphi & 0 & \cos \theta \cos \varphi \\
i \sin \varphi & \cos \theta \cos \varphi & 0
\end{array}\right]
$$

giving the dynamical modes and the associated phases

$$
\begin{gather*}
\left|\phi_{0}(t)\right\rangle=\left[\begin{array}{c}
\cos \theta \cos \varphi \\
-i \sin \varphi \\
-\sin \theta \cos \varphi
\end{array}\right], \quad \lambda_{0}=0, \quad \dot{\xi}_{0}=0,  \tag{2.39a}\\
\left|\phi_{ \pm}(t)\right\rangle=\frac{1}{\sqrt{2}}\left[\begin{array}{c}
\cos \theta \sin \varphi \pm i \sin \theta \\
i \cos \varphi \\
-\sin \theta \sin \varphi \pm i \cos \theta
\end{array}\right], \quad \lambda_{ \pm}= \pm \alpha_{0}, \quad \dot{\xi}_{ \pm}=\mp \frac{\dot{\theta}}{\sin \varphi} . \tag{2.39b}
\end{gather*}
$$

### 2.3 Single-mode driving: From adiabatic to exact passage

We consider the passage from the initial state $|1\rangle$ to the target state $|3\rangle$ in the three-state problem. Adiabatic passage assumes a slow evolution of the parameters such that adiabatic theorem applies: the dynamics is achieved approximately along a single adiabatic state, which connects the initial with the final states. In the three-state system, the considered adiabatic state is the well-known dark state

$$
\left|\phi_{D}(t)\right\rangle=\left[\begin{array}{c}
\cos \theta  \tag{2.40}\\
0 \\
-\sin \theta
\end{array}\right]
$$

where $\theta$ is the mixing angle, $\tan \theta=\Omega_{p} / \Omega_{s}$. We emphasize that the dynamical mode (2.39a) corresponds to the dark state for $\varphi=0$. The initial state $|1\rangle$ can in principle connect to the dark state in a counterintuitive sequence, i.e. when the Stokes precedes the pump pulse (i.e. $\theta$ going from 0 to $\pi / 2$ ), but the dynamics cannot permanently project exactly on it since the dark state is connected to the other adiabatic states through the non-adiabatic coupling. This coupling can be made small only for a slow variation of the parameters (adiabatic passage), which corresponds to a small value of $\varphi$. The Lewis-Riesenfeld invariant allows one to derive an alternative dynamical exact basis, on which one can project the dynamics exactly from the initial condition to a desired target state. The dark state can thus be interpreted as an approximation of the mode $\left|\phi_{0}(t)\right\rangle$. Accelerating the dynamics along this mode will necessarily induce population on the lossy excited state (through the component $-i \sin \varphi$ ).

One can for instance impose a single-invariant-mode driving by imposing at the initial time $\theta\left(t_{i}\right)=0$ and $\varphi\left(t_{i}\right)=0$, with the final boundary $\theta\left(t_{f}\right)=\pi / 2$ and $\varphi\left(t_{f}\right)=0$ at time $t_{f}$, which then drives the dynamics into the target state. These boundaries have to be taken into account in the definition of the pulses (2.37) to ensure the desired transfer. Such a single-mode dynamics can be interpreted as an exact passage compared to the
adiabatic passage occurring approximately along the dark state. Keeping the boundaries, we still have the freedom to choose any time dependent shape of the angles $\theta(t)$, $\varphi(t)$. For instance, the dynamics can also feature a speed up with respect to adiabatic passage or a certain degree of robustness by an appropriate time parametrization of the angles $\theta(t)$ and $\varphi(t)$. In Ref. [35], a recipe is provided to derive robust pulse in two-level systems. In Ref. [36], it is analyzed for $\Lambda$ systems. Specific optimizations can also guide the shaping as studied in the next chapters.

### 2.4 Multi-mode driving

One can determine other representations of the solutions, such as multi-mode solutions, which involve the three modes. We can choose for instance a multi-mode solution using $\theta\left(t_{i}\right)=0$, denoting the initial boundary $\varphi\left(t_{i}\right)=\varphi_{i}$, according to (2.5):

$$
\begin{equation*}
|\psi(t)\rangle=c_{0}\left|\phi_{0}(t)\right\rangle+c_{+} e^{i \xi_{+}(t)}\left|\phi_{+}(t)\right\rangle+c_{-} e^{i \xi_{-}(t)}\left|\phi_{-}(t)\right\rangle \tag{2.41}
\end{equation*}
$$

with

$$
\begin{align*}
c_{0} & =\left\langle\phi_{0}\left(t_{i}\right) \mid 1\right\rangle=\cos \varphi_{i},  \tag{2.42a}\\
c_{+} & =\left\langle\phi_{+}\left(t_{i}\right) \mid 1\right\rangle=\sin \varphi_{i} / \sqrt{2},  \tag{2.42b}\\
c_{-} & =\left\langle\phi_{-}\left(t_{i}\right) \mid 1\right\rangle=\sin \varphi_{i} / \sqrt{2}, \tag{2.42c}
\end{align*}
$$

and the Lewis-Riesenfeld phase $\xi_{0}=0\left(\right.$ since $\left.\lambda_{0}=0\right)$.
For simplicity we consider the simple parametrization with $t \in\left[t_{i} \equiv 0, t_{f} \equiv T\right]$ :

$$
\begin{equation*}
\varphi(t)=\varphi_{i}, \quad \theta(t)=\frac{\pi}{2 T} t \tag{2.43}
\end{equation*}
$$

giving the phase

$$
\begin{equation*}
\xi_{ \pm}(t)=\mp \xi(t)=\mp \frac{\theta(t)}{\sin \varphi_{i}} . \tag{2.44}
\end{equation*}
$$

The probability amplitude in state $|3\rangle$ at time $t$ reads

$$
\begin{equation*}
\langle 3 \mid \psi(t)\rangle=-\sin \theta(t)\left(\cos ^{2} \varphi_{i}+\cos \xi_{+}(t) \sin ^{2} \varphi_{i}\right)-\sin \varphi_{i} \cos \theta(t) \sin \xi_{+}(t), \tag{2.45}
\end{equation*}
$$

which gives at final time

$$
\begin{equation*}
\langle 3 \mid \psi(T)\rangle=-\left(\cos ^{2} \varphi_{i}+\cos \xi_{+}(T) \sin ^{2} \varphi_{i}\right) . \tag{2.46}
\end{equation*}
$$

The complete population transfer is then finally achieved when

$$
\begin{equation*}
\cos \xi_{+}(T)=\cos \left(\frac{\pi}{2 \sin \varphi_{i}}\right)=1 \tag{2.47}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
\sin \varphi_{i}=\frac{1}{4 k}, \quad k= \pm 1, \pm 2, \pm 3, \cdots \tag{2.48}
\end{equation*}
$$

The resulting pulses

$$
\begin{align*}
& \Omega_{p}=\frac{\pi}{T} \sqrt{16 k^{2}-1} \sin \left(\frac{\pi}{2 T} t\right)  \tag{2.49a}\\
& \Omega_{s}=\frac{\pi}{T} \sqrt{16 k^{2}-1} \cos \left(\frac{\pi}{2 T} t\right) \tag{2.49b}
\end{align*}
$$

feature a counterintuitive sequence. The smallest pulse energy required for the complete transfer with such pulses is for $k= \pm 1$.

An important quantity that will be considered frequently in this work is the time area of the transient population in the excited state:

$$
\begin{equation*}
A=\int_{0}^{T}|\langle 2 \mid \psi(t)\rangle|^{2} \tag{2.50}
\end{equation*}
$$

It will indeed characterize with a good approximation of the loss in the Raman system during the process through the lossy upper state, see Section 4.1. We determine its value for this multi-mode driving:

$$
\begin{equation*}
A=T \cos ^{2} \varphi_{i} \sin ^{2} \varphi_{i}\left[\frac{3}{2}+\frac{\sin \varphi_{i}}{\pi}\left(\frac{1}{2} \sin \frac{\pi}{\sin \varphi_{i}}-4 \sin \frac{\pi}{2 \sin \varphi_{i}}\right)\right] \tag{2.51}
\end{equation*}
$$

We show in Figure 2.2 the corresponding dynamics for $k=1$, giving $A \simeq 0.0879 T$, and in Figure 2.3 for $k=2$ giving $A \simeq 0.0231 T$. One can notice that the number of peaks of $P_{2}(t)$ is given by $k$.

One can remark that this dynamics with such pulses can be reinterpreted as a singlemode dynamics with different angles $\theta$ and $\varphi$ (and the required boundary conditions $\left.\theta\left(t_{i}\right)=0, \varphi\left(t_{i}\right)=0, \theta\left(t_{f}\right)=\pi / 2, \varphi\left(t_{f}\right)=0\right)$ given by

$$
\begin{align*}
\cos \varphi \cos \theta & =\left[1+\sin ^{2} \varphi_{i}(\cos \xi(t)-1)\right] \cos (\pi t / 2 T)+\sin \varphi_{i} \sin \xi(t) \sin (\pi t / 2 T)  \tag{2.52a}\\
\sin \varphi & =\cos \varphi_{i} \sin \varphi_{i}(1-\cos \xi(t))  \tag{2.52b}\\
\cos \varphi \sin \theta & =\left[1+\sin ^{2} \varphi_{i}(\cos \xi(t)-1)\right] \sin (\pi t / 2 T)-\sin \varphi_{i} \sin \xi(t) \cos (\pi t / 2 T) \tag{2.52c}
\end{align*}
$$

with (2.44): $\xi(t)=\theta(t) / \sin \varphi_{i}$. The curves of $\theta$ and $\varphi$ for $k=1$ are shown in Figure 2.4 .


Figure 2.2: Pump $\Omega_{p}$ (upper frame, solid red line) and Stokes $\Omega_{s}$ (upper frame, solid blue line) (in units of $1 / T)$ and the resulting populations with $\psi(t)$ the state solution (lower frame) for $k=1$.


Figure 2.3: Pump $\Omega_{p}$ (upper frame, solid red line) and Stokes $\Omega_{s}$ (upper frame, solid blue line) (in units of $1 / T$ ) and the resulting populations with $\psi(t)$ the state solution (lower frame) for $k=2$.


Figure 2.4: Time-dependence of the parameters $\varphi$ (upper frame, solid cyan line) and $\theta$ (lower frame, solid magenta line) (in units of $1 / \mathrm{T}$ ) determined from Eq. (2.52) for $k=1$.

### 2.5 Disscussion

In this chapter, we have shown that the Lewis-Riesenfeld invariant allows the derivation of a mode that can be interpreted as an exact version of the well-known dark state leading to adiabatic passage (STIRAP). This mode induces an exact transfer instead of adiabatic and approximate and has to be considered in an inverse engineering procedure: choosing the dynamics of the angles that generate it and taking the appropriate boundaries lead to the pulse shapes that ensure an exact transfer. This transfer is referred to as stimulated Raman exact passage (STIREP). We emphasize that, as expected, this mode that leads to an exact transfer features a non-zero transient component in the excited state contrary to the dark state.

Such transient population in the excited state is important to minimize since it will lead in practice to loss due to spontaneous emission. We expect typically counterintuitive pulse sequences (first Stokes, next pump) to do so. We have derived a simple (counterintuitive) sin / cos solution using a simple linear parametrization of the angles in a multi-mode configuration (that can be reinterpreted as a more complicated mono-mode solution). This solution is interesting since it is the opposite sequence of the one (intuitive $\cos / \sin$ ) that is the optimal solution minimizing the pulse energy $\mathcal{E}=\frac{\hbar}{4} \int_{0}^{T}\left(\Omega_{p}^{2}+\Omega_{s}^{2}\right) d s=\frac{3 \pi^{2}}{4} \frac{\hbar}{T}$ (see Section 3.6). In that case, the optimal intuitive sequence is thus fast (in fact the fastest one for given peak pulses) but leads to a large transient population in the excited state, which can be characterized by its time area: $A=\int_{0}^{T} \sin ^{2} \varphi(t) d t \simeq 0.3750 T$, and is thus subject to loss in practice. The counterintuitive $\sin$ / $\cos$ sequence that we have derived is slower (but exact) and requires five times more energy $\mathcal{E}=\frac{\hbar}{4} \int_{0}^{T}\left(\Omega_{p}^{2}+\Omega_{s}^{2}\right) d s=\frac{15 \pi^{2}}{4} \frac{\hbar}{T}$ but gives four times smaller population in the excited state $A \simeq 0.0879 T$. Optimization including losses is the subject of Chapter 4.

## Techniques of optimal control in quantum systems

## Contents

3.1 Introduction22
3.2 Lagrange multiplier method ..... 23
3.2.1 Description of the method ..... 23
3.2.2 Example ..... 25
3.2.3 Extension of the method for a time-dependent constraint ..... 27
3.3 Euler-Lagrange principle ..... 28
3.4 The Pontryagin maximum principle ..... 31
3.4.1 The Lagrangian problem with constraints and controls ..... 32
3.4.2 The PMP for the Mayer problem ..... 33
3.4.3 The PMP for the Mayer-Lagrange or Lagrange problems ..... 34
3.5 Optimal control with constrained controls ..... 36
3.6 Maximum Principle and shooting method for the three-level problem ..... 38
3.6.1 Original coordinates ..... 38
3.6.2 Spherical coordinates ..... 42
3.7 Optimal Control by gradient method ..... 48
3.8 Trigonometric expansion for finite-time stimulated Raman ex- act passage (STIREP) by gradient method ..... 56
3.8.1 Counterintuitive sequence as a starting point in a 8-parameter space ..... 56
3.8.2 Intuitive sequence ..... 61
3.8.3 Gradient method ..... 65

Optimal control problems of quantum systems can be outlined as seeking extremum solutions with respect to given costs. Generally, from a mathematical model represented typically by a differential equation, and additional constraints, one has first to formulate
appropriate objective functions which depend on actual demands such as the shortest control time, the smallest control field energy, or other optimal values of certain variables $[18,46]$. Optimal control techniques allow one to obtain optimal trajectories in the parameter space.

Section 3.1 gives the introduction and formulation of optimal control problems in the general form, of the Mayer-Lagrange type. Section 3.2 presents the Lagrange multiplier method for finding the extremums of functions with constraints. Section 3.3 presents the Euler-Lagrange principle for finding the optimal trajectory with fixed initial and final boundaries. Section 3.4 states the Pontryagin maximum principle (PMP), which is expressed according to three typical problems. In Section 3.5, we consider the optimization of constrained controls, used for instance for problems of time minimization. Section 3.6 describes the specific analysis of the techniques of optimal control theory applied to a three-level $\Lambda$-system. We present the gradient method in Section 3.7 to manage the optimal control by iterative algorithms and numerical analyses. Section 3.8 shows a practical example of application of the gradient method to the quantum control of $\Lambda$ systems.

### 3.1 Introduction

We consider the general control problem of the Mayer-Lagrange (or Bolza) type [18, 47]:

$$
\left\{\begin{array}{c}
J(x, u)=g\left(x\left(t_{f}\right), t_{f}\right)+\int_{t_{i}}^{t_{f}} f_{0}(x(t), u(t), t) d t,  \tag{3.1}\\
\dot{x}(t)=f(x(t), u(t), t) \\
x\left(t_{i}\right)=x_{i}
\end{array}\right.
$$

which consists in finding $m$ external controls $u(t) \in \mathbb{R}^{m}$ driving a dynamical system $\dot{x}(t)=f(x(t), u(t), t)$ of trajectory $x(t) \in \mathbb{R}^{n}$ with the initial condition $x\left(t_{i}\right)=x_{i}$, which minimizes the cost $J$, defined by a terminal condition $g\left(x\left(t_{f}\right), t_{f}\right)$, which describes a property of the state $x\left(t_{f}\right)$ at final time $t_{f}$, e.g. its distance from a chosen target state, and a chosen function $f_{0}$ taken into account via an integral:

$$
\begin{equation*}
u=\underset{v}{\arg \min }(J(x, v)) . \tag{3.2}
\end{equation*}
$$

The Mayer-Lagrange problem features a free final state in its general formulation.
When the final point of the trajectory is fixed, $x\left(t_{f}\right)=x_{f}$, one has to consider additionally $g\left(x\left(t_{f}\right), t_{f}\right)=0$ in the cost, which is referred to as a Lagrange problem [18].

On the other hand, the Mayer problem [18] consists in minimizing a terminal cost
(with a free final state): $J\left(x\left(t_{f}\right)\right)=g\left(x\left(t_{f}\right), t_{f}\right)$.
As an example, minimizing the energy corresponds to $f_{0}(x(t), u(t), t)=\eta\|u\|^{2}$, where $\eta>0$ expresses a compromise between the goal of transferring the state according to $g\left(x\left(t_{f}\right)\right)$ and the goal of keeping the energy of the controls small. Minimizing the time to reach a given target state from a given initial state corresponds to $f_{0}=1$.

A Mayer (or Mayer-Lagrange) problem can be transformed into a Lagrange problem and, conversely, a Lagrange problem into a Mayer problem (see below, in Section 3.4).

### 3.2 Lagrange multiplier method

### 3.2.1 Description of the method

The Lagrange multiplier method addresses the optimization problem of finding the extremum of a function $g(x), x \in \mathbb{R}^{n}$ under the constraint $f(x)=0$ (first assumed scalar for simplicity, i.e. $f \in \mathbb{R}$ ):

$$
\left\{\begin{array}{c}
J(x)=g(x),  \tag{3.3}\\
f(x)=0
\end{array}\right.
$$

In such problems, no dynamics is considered, time can be omitted. The extremum means that $\frac{\partial g}{\partial x_{i}}=0$ for all $x_{i}, i=1, \cdots, n$, if the $x_{i}$ are all independent. But the constraint introduces a dependence between the $x_{i}$ : one can express one $x_{j}$ as a function of the $n-1$ other $x_{i}$; choosing $j=n$ :

$$
\begin{equation*}
x_{n}=x_{n}\left(x_{1}, \cdots, x_{n-1}\right) \tag{3.4}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
d x_{n}=\frac{\partial x_{n}}{\partial x_{1}} d x_{1}+\cdots+\frac{\partial x_{n}}{\partial x_{n-1}} d x_{n-1} \tag{3.5}
\end{equation*}
$$

We plug this relation into $d g=\frac{\partial g}{\partial x_{1}} d x_{1}+\cdots+\frac{\partial g}{\partial x_{n}} d x_{n}$ :

$$
\begin{equation*}
d g=\left(\frac{\partial g}{\partial x_{1}}+\frac{\partial g}{\partial x_{n}} \frac{\partial x_{n}}{\partial x_{1}}\right) d x_{1}+\cdots+\left(\frac{\partial g}{\partial x_{n-1}}+\frac{\partial g}{\partial x_{n}} \frac{\partial x_{n}}{\partial x_{n-1}}\right) d x_{n-1} \tag{3.6}
\end{equation*}
$$

leading to

$$
\begin{equation*}
\frac{\partial g}{\partial x_{i}}+\frac{\partial g}{\partial x_{n}} \frac{\partial x_{n}}{\partial x_{i}}=0, \quad i=1, \cdots, n-1 \tag{3.7}
\end{equation*}
$$

since $g$ is extremum $d g=0$. We also have an equivalent relation for $f$ since $f=0$ :

$$
\begin{equation*}
\frac{\partial f}{\partial x_{i}}+\frac{\partial f}{\partial x_{n}} \frac{\partial x_{n}}{\partial x_{i}}=0, \quad i=1, \cdots, n-1 \tag{3.8}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
\frac{\partial x_{n}}{\partial x_{i}}=-\frac{\partial f}{\partial x_{i}}\left(\frac{\partial f}{\partial x_{n}}\right)^{-1} \tag{3.9}
\end{equation*}
$$

Combining with the expression (3.7) leads to

$$
\begin{equation*}
\frac{\partial g}{\partial x_{i}}-\frac{\partial g}{\partial x_{n}} \frac{\partial f}{\partial x_{i}}\left(\frac{\partial f}{\partial x_{n}}\right)^{-1}=0, \quad i=1, \cdots, n-1 \tag{3.10}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
\frac{\frac{\partial g}{\partial x_{i}}}{\frac{\partial f}{\partial x_{i}}}=\frac{\frac{\partial g}{\partial x_{n}}}{\frac{\partial f}{\partial x_{n}}}, \quad i=1, \cdots, n-1 \tag{3.11}
\end{equation*}
$$

We define the constant Lagrange multiplier ensuring the preceding equality (3.11) for all $x_{i}$ [48]:

$$
\begin{equation*}
\lambda=-\frac{\partial g}{\partial x_{n}}\left(\frac{\partial f}{\partial x_{n}}\right)^{-1}, \quad \text { i.e. } \quad \frac{\partial g}{\partial x_{n}}+\lambda \frac{\partial f}{\partial x_{n}}=0 \tag{3.12}
\end{equation*}
$$

The resulting equations that have to be solved read thus:

$$
\begin{equation*}
\frac{\partial g}{\partial x_{i}}+\lambda \frac{\partial f}{\partial x_{i}}=0, \quad i=1, \cdots, n \tag{3.13}
\end{equation*}
$$

One can reformulate it using a Lagrangian

$$
\begin{equation*}
L=g+\lambda f \tag{3.14}
\end{equation*}
$$

and one has to solve (3.13):

$$
\begin{equation*}
\frac{\partial L}{\partial x_{i}}=0, \quad \frac{\partial L}{\partial \lambda}=f=0 \tag{3.15}
\end{equation*}
$$

corresponding to solving

$$
\begin{equation*}
\frac{\partial L}{\partial q_{i}}=0, \quad i=1, \cdots, n+1 \tag{3.16}
\end{equation*}
$$

for the augmented system of vectorial coordinate $q=\left[x^{\top}, \lambda\right]^{\top}$.
If there are several constraints, i.e. $f$ is a vector, one has to consider a Lagrange multiplier $\lambda_{i}$ for each constraint, leading to a vector $\lambda$ of the same dimension of $f$. The Lagrangian reads then:

$$
\begin{equation*}
L=g+\lambda^{\top} f \tag{3.17}
\end{equation*}
$$

with the scalar product $\lambda^{\top} f=\sum_{i} \lambda_{i} f_{i}$ [48].

### 3.2.2 Example

As an interesting and simple example [49], we can consider the problem of finding the extremum of a quantum mean value $\langle A\rangle_{|\Psi\rangle}$ of an observable $A$ in a state

$$
\begin{equation*}
|\Psi\rangle=\sum_{i=1}^{N} a_{i}|i\rangle, \tag{3.18}
\end{equation*}
$$

expanded in the basis $\{|i\rangle, i=1, \cdots, N\}$. The constraint is the quantum normalization:

$$
\begin{equation*}
\sum_{i=1}^{N}\left|a_{i}\right|^{2}=1 \tag{3.19}
\end{equation*}
$$

We have thus to find the extremum of

$$
\begin{equation*}
g\left(a_{1}, a_{2}, \cdots, a_{N}, a_{1}^{*}, a_{2}^{*}, \cdots, a_{N}^{*}\right):=\langle A\rangle_{|\Psi\rangle}=\sum_{i, j} a_{i}^{*} a_{j}\langle i| A|j\rangle, \tag{3.20}
\end{equation*}
$$

i.e., the components $\left\{a_{i}\right\}$ of the state which lead to the extremum (minimum or maximum of $g$ ) under the constraint (3.19) which also reads

$$
\begin{equation*}
f:=\sum_{i=1}^{N} a_{i} a_{i}^{*}-1 \equiv 0 \tag{3.21}
\end{equation*}
$$

The Lagrange multiplier method shows the following equations according to (3.13):

$$
\begin{align*}
& \frac{\partial g}{\partial a_{n}^{*}}+\lambda \frac{\partial f}{\partial a_{n}^{*}}=0,  \tag{3.22a}\\
& \frac{\partial g}{\partial a_{n}}+\lambda \frac{\partial f}{\partial a_{n}}=0 \tag{3.22b}
\end{align*}
$$

in which, $\lambda$ is the Lagrange multiplier and the other components can be obtained from (3.20) and (3.21):

$$
\begin{align*}
\frac{\partial g}{\partial a_{n}^{*}} & =\sum_{j=1}^{N} a_{j}\langle n| A|j\rangle, & & \frac{\partial f}{\partial a_{n}^{*}}=a_{n}  \tag{3.23}\\
\frac{\partial g}{\partial a_{n}} & =\sum_{i=1}^{N} a_{i}^{*}\langle i| A|n\rangle, & & \frac{\partial f}{\partial a_{n}}=a_{n}^{*}
\end{align*}
$$

We remark that, in this problem, we have considered for convenience the two sets of variables $a_{i}, a_{i}^{*}$ instead of the real and imaginary parts of the $a_{i}$ 's. This is possible since
$\lambda$ is real as shown below (see Eq. (3.25)). Hence, (3.22) becomes

$$
\begin{align*}
& \sum_{i=1}^{N} a_{i}\langle n| A|i\rangle+\lambda a_{n}=0  \tag{3.24a}\\
& \sum_{i=1}^{N} a_{i}^{*}\langle i| A|n\rangle+\lambda a_{n}^{*}=0 \tag{3.24b}
\end{align*}
$$

that is, if $\lambda$ is real,

$$
A\left[\begin{array}{c}
a_{1}  \tag{3.25}\\
\vdots \\
a_{N}
\end{array}\right]=-\lambda\left[\begin{array}{c}
a_{1} \\
\vdots \\
a_{N}
\end{array}\right]
$$

which corresponds to an eigenvalue problem. Since $A$ is hermitian, the eigenvalues $\chi \equiv-\lambda$ are real, which is compatible with the passage from (3.24) to (3.25):

$$
A\left[\begin{array}{c}
a_{1}  \tag{3.26}\\
\vdots \\
a_{N}
\end{array}\right]=\chi\left[\begin{array}{c}
a_{1} \\
\vdots \\
a_{N}
\end{array}\right]
$$

We remark that, according to the method, all the eigenvalues give an extremum solution. Eq. (3.24) can be simply rewritten as:

$$
\begin{equation*}
\sum_{i=1}^{N} a_{i}\langle n| A|i\rangle=-\lambda a_{n} \equiv \chi a_{n} \tag{3.27}
\end{equation*}
$$

Plugging (3.27) into (3.20), we obtain

$$
\begin{align*}
\langle A\rangle_{|\Psi\rangle} & =\sum_{i} a_{i}^{*} \sum_{j} a_{j}\langle i| A|j\rangle \\
& =\sum_{i} a_{i}^{*} \chi a_{i}  \tag{3.28}\\
& =\chi \sum_{i}\left|a_{i}\right|^{2} \\
& =\chi
\end{align*}
$$

which means that the absolute maximum (minimum) value of a quantum mean value $\langle A\rangle_{|\Psi\rangle}$ is the maximum (minimum) value of the eigenvalues $\chi$ of $A$, realized by the state $|\Psi\rangle$ being the corresponding eigenstate. One concludes that applying the Lagrange multiplier method transforms the problem of finding the extremum of a quantum mean
value of an operator $A$ into the one of finding the extremum of the eigenvalue of $A$.

### 3.2.3 Extension of the method for a time-dependent constraint

If we consider the extremum of a function $g(x)$ with a time-dependent vectorial constraint of the type

$$
\begin{equation*}
f(x(t), \dot{x}(t), t)=0, \quad t \in\left[t_{i}, t_{f}\right] \tag{3.29}
\end{equation*}
$$

we have to introduce a Lagrange multiplier vector of the dimension of $f$ for each time $t$, i.e. the Lagrange multiplier vector becomes itself time dependent $\lambda(t)$. The scalar product in (3.17) has to be extended to time via an integral and the Lagrangian $\mathcal{L}$ becomes:

$$
\begin{equation*}
\mathcal{L}(x)=g(x)+\int_{t_{i}}^{t_{f}} \lambda^{\top}(t) f(x(t), \dot{x}(t), t) d t \tag{3.30}
\end{equation*}
$$

where $g(x)$ is defined for convenience through a functional:

$$
\begin{equation*}
g(x)=\int_{t_{i}}^{t_{f}} L_{0}(x(t), \dot{x}(t), t) d t \tag{3.31}
\end{equation*}
$$

We will consider typically the constraint as a first order differential equation:

$$
\begin{equation*}
\dot{x}=f(x(t), t) \tag{3.32}
\end{equation*}
$$

giving

$$
\begin{equation*}
\mathcal{L}(x)=\int_{t_{i}}^{t_{f}} L_{0}(x(t), \dot{x}(t), t) d t+\int_{t_{i}}^{t_{f}} \lambda^{\top}(t)\left(\frac{d}{d t} x-f(x(t), t)\right) d t \tag{3.33}
\end{equation*}
$$

Application of the method leads to

$$
\begin{equation*}
\frac{\partial \mathcal{L}}{\partial x}=0, \quad \frac{\partial \mathcal{L}}{\partial \lambda^{\top}}=0 \tag{3.34}
\end{equation*}
$$

The second equation allows one to recover the constraint $\dot{x}=f(x, t)$. The first equation has to be applied after an integration by part.

$$
\begin{equation*}
\mathcal{L}=\int_{t_{i}}^{t_{f}} d t L_{0}(x(t), \dot{x}(t), t)+\left[\lambda^{\top}(t) x(t)\right]_{t_{i}}^{t_{f}}-\int_{t_{i}}^{t_{f}} d t\left(\dot{\lambda}^{\top} x+\lambda^{\top}(t) f(x(t), t)\right) \tag{3.35}
\end{equation*}
$$

giving

$$
\begin{equation*}
\frac{\partial \mathcal{L}}{\partial x}=\int_{t_{i}}^{t_{f}} d t\left(\frac{\partial L_{0}}{\partial x}-\dot{\lambda}^{\top}-\lambda^{\top} \frac{\partial f}{\partial x}\right)=0 \tag{3.36}
\end{equation*}
$$

This gives the differential equations for the Lagrange multipliers, called costate [18]:

$$
\begin{equation*}
\dot{\lambda}^{\top}=\frac{\partial L_{0}}{\partial x}-\lambda^{\top} \frac{\partial f}{\partial x} . \tag{3.37}
\end{equation*}
$$

The problem introduced in Section 3.1 can be formulated in similar way. We consider the dynamical problem (the constraint):

$$
\begin{equation*}
\dot{x}=f(x(t), u(t), t), \quad x\left(t_{i}\right)=x_{i} \tag{3.38}
\end{equation*}
$$

with $m$ control variables $u(t)$ minimizing a cost

$$
\begin{equation*}
J(x, u)=g_{f}+\int_{t_{i}}^{t_{f}} f_{0}(x(t), u(t), t) d t \tag{3.39}
\end{equation*}
$$

with $g_{f}$ a terminal condition and $f_{0}$ a given function. In addition to the equation (3.34), we have to consider

$$
\begin{equation*}
\frac{\partial \mathcal{L}}{\partial u}=0, \tag{3.40}
\end{equation*}
$$

where $\mathcal{L}$ now writes:

$$
\begin{equation*}
\mathcal{L}(x, u)=g_{f}+\int_{t_{i}}^{t_{f}} f_{0}(x(t), u(t), t) d t+\int_{t_{i}}^{t_{f}} \lambda^{\top}(t)(\dot{x}-f(x(t), u(t), t)) d t, \tag{3.41}
\end{equation*}
$$

which gives

$$
\begin{equation*}
\frac{\partial f_{0}}{\partial u}-\lambda^{\top}(t) \frac{\partial f}{\partial u}=0, \tag{3.42}
\end{equation*}
$$

which links the controls $u$ to the costate $\lambda$. As formulated, the problem is difficult to solve since it needs to solve the differential (3.37) for the costate, which is in general at least of the same complexity of the original dynamical equation; for all the possible $u(t)$. An approximate iterative algorithm is needed in general to find a local extremum and we have no guarantee that this extremum will be a (global) optimum. The alternative formulation using the Pontryagin maximum principle (see Section 3.4) will allow one to find exact solutions for relatively simple problems.

### 3.3 Euler-Lagrange principle

The Lagrangian multiplier method for a time-dependent problem can be in fact reformulated and generalized with the Euler-Lagrange principle. The Euler-Lagrange principle corresponds to finding the trajectory $x(t)$ of fixed boundaries $x\left(t_{i}\right)=x_{i}$ and
$x\left(t_{f}\right)=x_{f}$, which realizes the extremum of the integral, defining the cost:

$$
\begin{equation*}
J(x)=\int_{t_{i}}^{t_{f}} L_{0}(x(t), \dot{x}(t), t) d t \tag{3.43}
\end{equation*}
$$

where we recognize $L_{0}$ as a Lagrangian and $J$ the corresponding action in classical mechanics. The integral of the cost of (3.1) takes precisely this latter form. The EulerLagrange principle can be expressed by the Euler-Lagrange equations [50]

$$
\begin{equation*}
\frac{d}{d t} \frac{\partial L_{0}}{\partial \dot{x}_{i}}=\frac{\partial L_{0}}{\partial x_{i}} \tag{3.44}
\end{equation*}
$$

which generalizes (3.14) to the dynamical case. The extremum trajectory $x(t)$ is solution of the Lagrange equation (3.44), or equivalently of the Hamilton equations

$$
\begin{equation*}
\dot{x}_{i}=\frac{\partial H_{0}}{\partial p_{i}}, \quad \dot{p}_{i}=-\frac{\partial H_{0}}{\partial x_{i}} \tag{3.45}
\end{equation*}
$$

with the conjugate moment

$$
\begin{equation*}
p_{i}=\frac{\partial L_{0}}{\partial \dot{x}_{i}} \tag{3.46}
\end{equation*}
$$

and the Hamiltonian (with $p$ a row vector)

$$
\begin{equation*}
H_{0}=p \dot{x}-L_{0} . \tag{3.47}
\end{equation*}
$$

If the system is subject to a (vectorial) constraint (of the same type of the one in Lagrange multiplier method)

$$
\begin{equation*}
f(x(t), \dot{x}(t), t)=0, \tag{3.48}
\end{equation*}
$$

one has to consider the augmented problem of coordinates $q(t)=\left[x(t)^{\boldsymbol{\top}}, \lambda^{\top}\right]^{\top}$, which features the modified Lagrangian:

$$
\begin{equation*}
L_{\lambda}=L_{0}(x(t), \dot{x}(t), t)+\lambda^{\top} f(x(t), \dot{x}(t), t), \tag{3.49}
\end{equation*}
$$

where the constraint is included via a real vector function $\lambda=\lambda(t)$ of Lagrange multipliers having the same dimension of $f$, and called the costate in this context. We notice that we recover Eq. (3.30) of Subsection 3.2.3 with the connection $\mathcal{L}(x)=\int_{t_{f}}^{t_{i}} L_{0}(x(t), \dot{x}(t), t) d t$. The Euler-Lagrange equations that have to be solved

$$
\begin{equation*}
\frac{d}{d t} \frac{\partial L_{\lambda}}{\partial \dot{q}_{i}}=\frac{\partial L_{\lambda}}{\partial q_{i}}, \tag{3.50}
\end{equation*}
$$

give for the Lagrange multiplier part

$$
\begin{equation*}
\frac{d}{d t} \frac{\partial L_{\lambda}}{\partial \dot{\lambda}}=0=\frac{\partial L_{\lambda}}{\partial \lambda}=f \tag{3.51}
\end{equation*}
$$

meaning that the constraint $f=0$ is satisfied, and for the dynamical variables

$$
\begin{equation*}
\frac{d}{d t} \frac{\partial L_{0}}{\partial \dot{x}_{i}}+\lambda^{\top} \frac{d}{d t} \frac{\partial f}{\partial \dot{x}_{i}}=\frac{\partial L_{0}}{\partial x_{i}}+\lambda^{\top} \frac{\partial f}{\partial x_{i}} . \tag{3.52}
\end{equation*}
$$

The latter can be rewritten as

$$
\begin{equation*}
\operatorname{grad} J(x)+\lambda \operatorname{grad} F(x)=0 \tag{3.53}
\end{equation*}
$$

with the general definition of the gradient:

$$
\begin{equation*}
\operatorname{grad} \Phi(x)=\frac{\partial \varphi}{\partial x}-\frac{d}{d t} \frac{\partial \varphi}{\partial \dot{x}}, \tag{3.54}
\end{equation*}
$$

for $\Phi(x)$ defined as

$$
\begin{equation*}
\Phi(x)=\int_{t_{i}}^{t_{f}} \varphi(x, \dot{x}, t) d t \tag{3.55}
\end{equation*}
$$

Here we have thus

$$
\begin{equation*}
F(x)=\int_{t_{i}}^{t_{f}} f(x, \dot{x}, t) d t \tag{3.56}
\end{equation*}
$$

The Euler-Lagrange method generalizes thus the Lagrange multiplier method to constraints and costs defined as functionals for dynamical problems.

If the constraint is a first order differential equation (with $f$ having the same dimension as $x$ )

$$
\begin{equation*}
\dot{x}=f(x, t), \tag{3.57}
\end{equation*}
$$

the Lagrangian becomes

$$
\begin{equation*}
L_{\lambda}=L_{0}(x, t)+\lambda^{\top}(\dot{x}-f) \tag{3.58}
\end{equation*}
$$

where $L_{0}(x, t)$ becomes only a function of $x$ and $t$, and the equations to solve are given by the Euler-Lagrange equations

$$
\begin{equation*}
\frac{d}{d t} \frac{\partial L_{\lambda}}{\partial \dot{q}_{i}}=\frac{\partial L_{\lambda}}{\partial q_{i}} \tag{3.59}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
0=\frac{\partial L_{\lambda}}{\partial \lambda}=\dot{x}-f \tag{3.60}
\end{equation*}
$$

meaning that the constraint $\dot{x}=f$ is satisfied, and

$$
\begin{equation*}
\frac{d}{d t} \frac{\partial L_{\lambda}}{\partial \dot{x}}=\dot{\lambda}^{\top}=\frac{\partial L_{\lambda}}{\partial x}=\frac{\partial L_{0}}{\partial x}-\lambda^{\top} \frac{\partial f}{\partial x}, \tag{3.61}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
\dot{\lambda}^{\top}=\frac{\partial L_{0}}{\partial x}-\lambda^{\top} \frac{\partial f}{\partial x}, \tag{3.62}
\end{equation*}
$$

which represents the adjoint equation (of the costate). We recover Eq. (3.37).
The problem can be reformulated by Hamilton equations with an Hamiltonian. The conjugate moments are by definition

$$
\begin{equation*}
p_{x}=\frac{\partial L_{\lambda}}{\partial \dot{x}}=\lambda^{\top}, \quad p_{\lambda}=\frac{\partial L_{\lambda}}{\partial \dot{\lambda}}=0 \tag{3.63}
\end{equation*}
$$

with the corresponding Hamiltonian

$$
\begin{equation*}
H=\lambda^{\top} \dot{x}-L_{\lambda}=\lambda^{\top} f-L_{0}, \tag{3.64}
\end{equation*}
$$

and the Hamilton equations

$$
\begin{equation*}
\dot{x}=\frac{\partial H}{\partial p_{x}}=f, \quad \dot{p}_{x}=\dot{\lambda}^{\top}=-\frac{\partial H}{\partial x}=\frac{\partial L_{0}}{\partial x}-\lambda^{\top} \frac{\partial f}{\partial x} . \tag{3.65}
\end{equation*}
$$

This shows that the conjugate moments are zero except the one associated to $x$, which is the Lagrange multiplier $p_{x}=\lambda^{\top}$ and solution of Eq. (3.62).

We remark that in this notation, the term $\frac{\partial f}{\partial x}$ corresponds to a matrix, which reads for instance for $n=2$ :

$$
\frac{\partial f}{\partial x}=\left[\begin{array}{ll}
\frac{\partial f_{1}}{\partial x_{1}} & \frac{\partial f_{1}}{\partial x_{2}}  \tag{3.66}\\
\frac{\partial f_{2}}{\partial x_{1}} & \frac{\partial f_{2}}{\partial x_{2}}
\end{array}\right]
$$

while $\frac{\partial L_{0}}{\partial x}$ is a row vector, i.e. for $n=2$ :

$$
\begin{equation*}
\frac{\partial L_{0}}{\partial x}=\left[\frac{\partial L_{0}}{\partial x_{1}}, \frac{\partial L_{0}}{\partial x_{2}}\right] . \tag{3.67}
\end{equation*}
$$

The problem is most often formulated with additional controls $u(t)$, as in Eq. (3.1), and solved using the Pontryagin maximum principle [17] as shown below.

### 3.4 The Pontryagin maximum principle

In this section, we follow the presentation of Ref. [18].

### 3.4.1 The Lagrangian problem with constraints and controls

We consider the control problem (3.1). Following [18], we connect the Lagrangian $L_{0}$ with $f_{0}$ as

$$
\begin{equation*}
L_{0}(x(t), u(t), t)=p_{0} f_{0}(x(t), u(t), t), \tag{3.68}
\end{equation*}
$$

with $p_{0}$ a constant. The coordinates of the augmented problem are here $q(t)=$ $\left[x(t)^{\top}, u(t)^{\top}, \lambda^{\top}\right]^{\top}$; the equations of the dynamics are considered as the constraints, and one remarks that the Lagrangian does not depend on $\dot{q}$. If $\{x, u\}$ is an optimal solution of (3.1), then there exists a vector function $\lambda=\lambda(t)$ of Lagrange multipliers such that $\{x, u, \lambda\}$ is an optimal for the unconstrained problem of minimizing

$$
\begin{equation*}
L_{\lambda}=L_{0}+\lambda^{\top}(\dot{x}(t)-f(x(t), u(t), t)) \tag{3.69}
\end{equation*}
$$

The Euler equations read

$$
\begin{align*}
& \frac{d}{d t} \frac{\partial L_{\lambda}}{\partial \dot{x}}=\dot{\lambda}^{\top}=\frac{\partial L_{\lambda}}{\partial x}=\frac{\partial L_{0}}{\partial x}-\lambda^{\top} \frac{\partial f}{\partial x},  \tag{3.70a}\\
& \frac{d}{d t} \frac{\partial L_{\lambda}}{\partial \dot{u}}=0=\frac{\partial L_{\lambda}}{\partial u}=\frac{\partial L_{0}}{\partial u}-\lambda^{\top} \frac{\partial f}{\partial u},  \tag{3.70b}\\
& \frac{d}{d t} \frac{\partial L_{\lambda}}{\partial \dot{\lambda}}=0=\frac{\partial L_{\lambda}}{\partial \lambda}=\dot{x}-f, \tag{3.70c}
\end{align*}
$$

i.e., as before,

$$
\begin{equation*}
\dot{\lambda}^{\top}=\frac{\partial L_{0}}{\partial x}-\lambda^{\top} \frac{\partial f}{\partial x}, \tag{3.71}
\end{equation*}
$$

which represents the adjoint equation (of the costate), and

$$
\begin{equation*}
\frac{\partial L_{0}}{\partial u}=\lambda^{\top} \frac{\partial f}{\partial u}, \tag{3.72}
\end{equation*}
$$

which links $u$ and $\lambda$. We notice that we recover Eq. (3.42) when $p_{0}=1$.
The conjugate moments are by definition

$$
\begin{equation*}
p_{x}=\frac{\partial L_{\lambda}}{\partial \dot{x}}=\lambda^{\top}, \quad p_{u}=\frac{\partial L_{\lambda}}{\partial \dot{u}}=0, \quad p_{\lambda}=\frac{\partial L_{\lambda}}{\partial \dot{\lambda}}=0 \tag{3.73}
\end{equation*}
$$

with the corresponding Hamiltonian

$$
\begin{equation*}
H=\lambda^{\top} \dot{x}-L_{\lambda}=\lambda^{\top} f-L_{0}=\lambda^{\top} f-p_{0} f_{0}, \tag{3.74}
\end{equation*}
$$

and the Hamilton equations

$$
\begin{equation*}
\dot{x}=\frac{\partial H}{\partial p_{x}}=f, \quad \dot{p}_{x}=\dot{\lambda}^{\top}=-\frac{\partial H}{\partial x}=p_{0} \frac{\partial f_{0}}{\partial x}-\lambda^{\top} \frac{\partial f}{\partial x} . \tag{3.75}
\end{equation*}
$$

This shows that, as before, the conjugate moments are zero except the one associated to $x$ which is the Lagrange multiplier $p_{x}=\lambda^{\top}$ and solution of Eq. (3.71). The problem is solved using the Pontryagin maximum principle as shown below.

Remark: Multiplying the Hamiltonian by a constant $\alpha, \alpha H=\alpha p_{x} f-\alpha p_{0} f_{0}$, comes down to consider the couple $\left(\alpha p_{x}, \alpha p_{0}\right)$, and leads to the same trajectory since $\dot{x}=$ $\frac{\partial \alpha H}{\partial \alpha p_{x}}=\frac{\partial H}{\partial p_{x}},\left(\alpha \dot{p}_{x}\right)=\alpha \dot{p}_{x}=-\frac{\partial \alpha H}{\partial x}=-\alpha \frac{\partial H}{\partial x}$.

### 3.4.2 The PMP for the Mayer problem

We first formulate the Pontryagin maximum principle (PMP) on the Mayer problem [of free final state and a fixed final time is also considered: $J\left(x\left(t_{f}\right)\right)=g\left(x\left(t_{f}\right)\right)$ ] [18], i.e. $f_{0}=0$ and, the adjoint equations of the costate and the Hamiltonian, read respectively:

$$
\begin{equation*}
\dot{\lambda}^{\top}=-\lambda^{\top} \frac{\partial f}{\partial x}, \quad H=\lambda^{\top} f \tag{3.76}
\end{equation*}
$$

The PMP states that the Hamiltonian $H$ is maximum (necessary conditions of optimality) [17], i.e.

$$
\begin{equation*}
\frac{\partial H}{\partial u}=\frac{\partial\left(\lambda^{\top} f(x, u)\right)}{\partial u}=\lambda^{\top} \frac{\partial f(x, u)}{\partial u}=0 \tag{3.77}
\end{equation*}
$$

for $u$ satisfying the adjoint equation (3.76) with the terminal condition

$$
\begin{equation*}
\lambda^{\top}\left(t_{f}\right)=-\frac{\partial g}{\partial x}\left(x\left(t_{f}\right)\right) \tag{3.78}
\end{equation*}
$$

when there is no bound on the controls $u$.
To prove (3.77), we analyse a small variation of the control: $u_{\varepsilon}(t):=u(t)+\varepsilon v(t)$ and denote $x_{\varepsilon}(t)$ the trajectory corresponding to $u_{\varepsilon}(t)$. The corresponding variation of the cost reads

$$
\begin{equation*}
\Delta J:=J\left(x_{\varepsilon}\left(t_{f}\right)\right)-J\left(x\left(t_{f}\right)\right) \approx \frac{\partial g}{\partial x}\left(x\left(t_{f}\right)\right)\left(x_{\varepsilon}\left(t_{f}\right)-x\left(t_{f}\right)\right) \tag{3.79}
\end{equation*}
$$

which we can write as

$$
\begin{equation*}
\Delta J \approx \frac{\partial g}{\partial x}\left(x\left(t_{f}\right)\right) z\left(t_{f}\right) \varepsilon=-\lambda^{\top}\left(t_{f}\right) z\left(t_{f}\right) \varepsilon \tag{3.80}
\end{equation*}
$$

using the definition

$$
\begin{equation*}
z(t):=\frac{d x_{\varepsilon}(t)}{d \varepsilon}(\varepsilon=0) \tag{3.81}
\end{equation*}
$$

and the terminal condition (3.78). Differentiating with respect to $t$ the latter allows one
to derive a differential equation for $z$ :

$$
\begin{align*}
\dot{z}(t)=\frac{d \dot{x}_{\varepsilon}(t)}{d \varepsilon}(\varepsilon=0) & =\frac{d f\left(x_{\varepsilon}(t), u(t)+\varepsilon v(t)\right)}{d \varepsilon}(\varepsilon=0) \\
& =\frac{\partial f(x(t), u(t))}{\partial x} z+\frac{\partial f(x(t), u(t))}{\partial u} v \tag{3.82}
\end{align*}
$$

with initial condition $z\left(t_{i}\right)=0$. Considering the adjoint equation

$$
\begin{equation*}
\dot{\lambda}^{\top}=-\lambda^{\top}(t) \frac{\partial f(x(t), u(t))}{\partial x}, \tag{3.83}
\end{equation*}
$$

and the writing (since $\left.z\left(t_{i}\right)=0\right)$ : $\lambda^{\top}\left(t_{f}\right) z\left(t_{f}\right)=\int_{t_{i}}^{t_{f}} \frac{d}{d t}\left(\lambda^{\top}(t) z(t)\right) d t=\int_{t_{i}}^{t_{f}}\left(\dot{\lambda}^{\top} z(t)+\right.$ $\left.\lambda^{\top}(t) \dot{z}\right) d t$, Eq. (3.80) can be rewritten as

$$
\begin{equation*}
\Delta J \approx-\varepsilon \int_{t_{i}}^{t_{f}} \lambda^{\top}(t) \frac{\partial f(x(t), u(t))}{\partial u} v(t) d t . \tag{3.84}
\end{equation*}
$$

The necessary condition for $u$ being a local minimum for $J$, i.e. $\Delta J=0$, corresponding to a local maximum for $\lambda^{\top}(t) f(x(t), u(t))$, i.e. the Hamiltonian being maximum, requires Eq. (3.77).

### 3.4.3 The PMP for the Mayer-Lagrange or Lagrange problems

If one has a Mayer-Lagrange (or Lagrange) problem instead of a Mayer problem, the conditions of the maximum principle can be adapted by transforming the problem into a Mayer problem. This is done by defining the extra (scalar) component $y(t)$, which satisfies the constraint:

$$
\begin{equation*}
\dot{y}=f_{0}(x, u, t), \quad y\left(t_{i}\right)=0 . \tag{3.85}
\end{equation*}
$$

Using this extra variable the cost takes indeed the Mayer form

$$
\begin{equation*}
J(u)=g\left(x\left(t_{f}\right), t_{f}\right)+y\left(t_{f}\right) . \tag{3.86}
\end{equation*}
$$

The costate now has dimension $n+1$. If we call $\lambda$ the first $n$ components of the costate and $\mu$ the remaining component, the adjoint equations take the form

$$
\begin{align*}
\dot{\lambda}^{\top} & =-\lambda^{\top}(t) \frac{\partial f(x(t), u(t))}{\partial x}-\mu \frac{\partial f_{0}(x(t), u(t))}{\partial x},  \tag{3.87a}\\
\dot{\mu} & =0 \tag{3.87b}
\end{align*}
$$

with the additional terminal condition

$$
\begin{equation*}
\mu\left(t_{f}\right)=-\frac{\partial\left(g\left(x, t_{f}\right)+y\right)}{\partial y}\left(y\left(t_{f}\right)\right)=-1 \tag{3.88}
\end{equation*}
$$

This leads to the Hamiltonian

$$
\begin{equation*}
H=\lambda^{\top} f+\mu f_{0}=\lambda^{\top} f-f_{0} \tag{3.89}
\end{equation*}
$$

which is maximum. As stated before, one can always for convenience consider the Hamiltonian multiplied by a (positive) constant $\alpha, \alpha H=\alpha \lambda^{\top} f-\alpha f_{0}$ with the same trajectories $x$ and $\lambda$. This means that we can always multiply $f_{0}$ by a positive constant $\alpha$ in the definition of the Hamiltonian (3.89). This constant $\alpha$ changes the $\lambda$ (as a multiplicative factor) but not the trajectory $x(t)$, neither the optimum $u$.

To summarize, in order to derive solutions that satisfy the necessary conditions of optimality, we have to solve the system of equations

$$
\begin{align*}
\dot{\lambda}^{\top} & =-\lambda^{\top}(t) \frac{\partial f(x(t), u(t))}{\partial x}+\frac{\partial f_{0}(x(t), u(t))}{\partial x}  \tag{3.90a}\\
\dot{x} & =f(x(t), u(t)) \tag{3.90b}
\end{align*}
$$

with the boundary conditions

$$
\begin{align*}
x\left(t_{i}\right) & =x_{i}  \tag{3.91a}\\
\lambda^{\top}\left(t_{f}\right) & =-\frac{\partial g}{\partial x}\left(x\left(t_{f}\right)\right) \tag{3.91b}
\end{align*}
$$

such that the Hamiltonian

$$
\begin{equation*}
H=\lambda^{\top} f+\mu f_{0}=\lambda^{\top} f-f_{0} \tag{3.92}
\end{equation*}
$$

is maximum, i.e.

$$
\begin{equation*}
\frac{\partial H}{\partial u}=0 . \tag{3.93}
\end{equation*}
$$

This is a two points boundary value problem as the boundary conditions are at time $t_{i}$ and $t_{f}$. The procedure of resolution is typically as follows: one leaves the initial condition for $\lambda, \lambda\left(t_{i}\right)$, as a free parameter, solves the equations (3.90) with the initial condition (3.91a), and then tries to adjust the value of the parameter $\lambda\left(t_{i}\right)$ so as to meet condition (3.91b). A typical method to implement this procedure is the shooting method (see below).

Every control $u$ that is obtained with this procedure satisfies the necessary conditions
of optimality and it is a candidate to be the optimal control. It is thus called an extremal. By comparing the costs $J$ obtained with the various extremals one finds the minimum cost and the corresponding optimal control.

For some relatively simple problems (see below), we will be able to determine the optimum exactly.

For a Lagrange problem, i.e. featuring a fixed end point $x\left(t_{f}\right)=x_{f}$, the boundary conditions become

$$
\begin{align*}
x\left(t_{i}\right) & =x_{i}  \tag{3.94a}\\
x\left(t_{f}\right) & =x_{f} \tag{3.94b}
\end{align*}
$$

In what follows, we consider quantum control problem associated the Schrödinger equation and the Hamiltonian of the driven quantum system. In order to distinguish the latter with the (classical) Hamiltonian resulting from the optimization procedure, we will refer to the latter as the pseudo Hamiltonian or the control Hamiltonian, denoted $H_{c}$.

### 3.5 Optimal control with constrained controls

We consider a pesudo (or control) Hamiltonian of the bilinear form [51]:

$$
\begin{equation*}
H_{c}=H_{0}+u_{1} H_{1}+u_{2} H_{2} \tag{3.95}
\end{equation*}
$$

where $H_{0}, H_{1}, H_{2}$ are independent of the controls $u_{1}, u_{2}$, and a constraint on the controls:

$$
\begin{equation*}
u_{1}^{2}+u_{2}^{2} \leq u_{0}^{2} \tag{3.96}
\end{equation*}
$$

The goal is to maximize $H_{c}$ for any value of $H_{0}, H_{1}, H_{2}$, under the above constraint. The maximization of $H_{c}$ corresponds to the necessary conditions $\frac{\partial H_{c}}{\partial u_{1}}=0, \frac{\partial H_{c}}{\partial u_{2}}=0$ according to the PMP, in the case of absence of constraint on the controls, but this is not true anymore with a constraint.

Keeping the full generality of the problem, we can rewrite the controls as follows:

$$
\begin{align*}
& u_{1}(t)=u_{m}(t) \cos (\theta(t))  \tag{3.97a}\\
& u_{2}(t)=u_{m}(t) \sin (\theta(t)) \tag{3.97b}
\end{align*}
$$

implying

$$
\begin{equation*}
u_{1}^{2}(t)+u_{2}^{2}(t)=u_{m}^{2}(t) \leq u_{0}^{2} \tag{3.98}
\end{equation*}
$$

This means that the constraint condition (3.96) is transferred to the condition (3.98) on $u_{m}: u_{m}^{2} \leq u_{0}^{2}$, which is independent of $\theta$. The maximization of $H_{c}$ corresponds thus to the necessary condition

$$
\begin{equation*}
\frac{\partial H_{c}}{\partial \theta}=0, \tag{3.99}
\end{equation*}
$$

which gives

$$
\begin{equation*}
\frac{\partial H_{c}}{\partial \theta}=\frac{\partial u_{1}}{\partial \theta} H_{1}+\frac{\partial u_{2}}{\partial \theta} H_{2}=u_{m}\left(-H_{1} \sin \theta+H_{2} \cos \theta\right)=0 \tag{3.100}
\end{equation*}
$$

ie.

$$
\begin{equation*}
\frac{H_{2}}{H_{1}}=\frac{\sin \theta}{\cos \theta} \tag{3.101}
\end{equation*}
$$

or

$$
\begin{align*}
& \cos \theta=\frac{H_{1}}{\sqrt{H_{1}^{2}+H_{2}^{2}}},  \tag{3.102a}\\
& \sin \theta=\frac{H_{2}}{\sqrt{H_{1}^{2}+H_{2}^{2}}}, \tag{3.102b}
\end{align*}
$$

which satisfy

$$
\begin{equation*}
\cos ^{2} \theta+\sin ^{2} \theta=1 \tag{3.103}
\end{equation*}
$$

and give

$$
\begin{align*}
& u_{1}=u_{m} \frac{H_{1}}{\sqrt{H_{1}^{2}+H_{2}^{2}}},  \tag{3.104a}\\
& u_{2}=u_{m} \frac{H_{2}}{\sqrt{H_{1}^{2}+H_{2}^{2}}} . \tag{3.104b}
\end{align*}
$$

We plug them in the pseudo Hamiltonian:

$$
\begin{equation*}
H_{c}=H_{0}+u_{m} \sqrt{H_{1}^{2}+H_{2}^{2}} . \tag{3.105}
\end{equation*}
$$

The pseudo Hamiltonian is then maximum when $u_{m}$ takes its maximum value, ie, according to (3.96)

$$
\begin{equation*}
u_{m}=u_{0} \tag{3.106}
\end{equation*}
$$

and it finally reads

$$
\begin{equation*}
H_{c}=H_{0}+u_{0} \sqrt{H_{1}^{2}+H_{2}^{2}} \tag{3.107}
\end{equation*}
$$

with the controls

$$
\begin{align*}
& u_{1}=u_{0} \frac{H_{1}}{\sqrt{H_{1}^{2}+H_{2}^{2}}}  \tag{3.108a}\\
& u_{2}=u_{0} \frac{H_{2}}{\sqrt{H_{1}^{2}+H_{2}^{2}}} \tag{3.108b}
\end{align*}
$$

One can remark that the controls attain the maximum of the constraint at each time:

$$
\begin{equation*}
u_{1}^{2}(t)+u_{2}^{2}(t)=u_{0}^{2} \tag{3.109}
\end{equation*}
$$

### 3.6 Maximum Principle and shooting method for the threelevel problem

We consider a three-level system Eq. (2.31) of Hamiltonian

$$
H(t)=\frac{1}{2}\left[\begin{array}{ccc}
0 & \Omega_{p}(t) & 0  \tag{3.110}\\
\Omega_{p}(t) & 0 & \Omega_{s}(t) \\
0 & \Omega_{s}(t) & 0
\end{array}\right]
$$

where $\Omega_{p}$ and $\Omega_{s}$ are the (real) Rabi frequencies describing the resonant interactions with the pump and the Stokes pulses. In this resonantly driven three-level system, the optimal pulses leading to the transfer are well known [20] (see below in spherical coordinates). We also rederive the result using the calculation in the original coordinates.

### 3.6.1 Original coordinates

Optimality can be analyzed in terms of pulse energy, i.e.

$$
\begin{equation*}
\mathcal{E}=\frac{1}{4} \int_{t_{i}}^{t_{f}}\left(\Omega_{p}^{2}(t)+\Omega_{s}^{2}(t)\right) d t \tag{3.111}
\end{equation*}
$$

or in terms of fluence (or generalized pulse area), i.e.

$$
\begin{equation*}
\mathcal{F}=\int_{t_{i}}^{t_{f}} \sqrt{\Omega_{p}^{2}(t)+\Omega_{s}^{2}(t)} d t \tag{3.112}
\end{equation*}
$$

Minimizing the functional $\mathcal{E}$ is equivalent to minimizing the transfer-time for controls bounded. The cost $\mathcal{F}$ is time-reparametrization invariant: one can always reparametrize the time of the optimal solution in order to obtain controls of lower amplitude. The optimal solutions corresponding to the two costs lead to the same trajectories in the
phase space [20].
In order to formulate the problem, one use the fact that this resonant three-state problem is of $\mathrm{SU}(2)$ symmetry. One can separate the dynamics into two independent sub-dynamics each of them being associated to the real and imaginary parts of the initial condition [20]. We consider for simplicity $\left\langle 1 \mid \psi\left(t_{i}\right)\right\rangle=1$, leading to the system of equations

$$
\begin{equation*}
\dot{\mathbf{x}}=\frac{1}{2}\left(\Omega_{p} T_{1}+\Omega_{s} T_{2}\right) \mathbf{x} \tag{3.113}
\end{equation*}
$$

with

$$
T_{1}=\left[\begin{array}{ccc}
0 & -1 & 0  \tag{3.114}\\
1 & 0 & 0 \\
0 & 0 & 0
\end{array}\right], \quad T_{2}=\left[\begin{array}{ccc}
0 & 0 & 0 \\
0 & 0 & -1 \\
0 & 1 & 0
\end{array}\right], \quad T_{3}=\left[\begin{array}{ccc}
0 & 0 & 1 \\
0 & 0 & 0 \\
-1 & 0 & 0
\end{array}\right]
$$

$\left[T_{1}, T_{2}\right]=T_{3}$, and $\mathbf{x}=\left[x_{1} \equiv\langle 1 \mid \psi\rangle, x_{2} \equiv i\langle 2 \mid \psi\rangle, x_{3} \equiv-\langle 3 \mid \psi\rangle\right]^{\top}$, where the superscript T denotes the transpose of the vector.

To determine optimal control fields $\mathbf{u}(t)$ of a dynamical system $\dot{\mathbf{x}}=\mathbf{f}(\mathbf{x}(t) ; \mathbf{u}(t)$ ) (of dimension $N$ ) with respect to the minimization of a given cost

$$
\begin{equation*}
J(\mathbf{u}(t))=\int_{t_{i}}^{t_{f}} g(\mathbf{x}(t), \mathbf{u}(t)) d t \tag{3.115}
\end{equation*}
$$

we use Pontryagin's maximum principle, which provides necessary conditions for optimality [17]. The maximum principle states that the trajectories of the extremal vector $\mathbf{x}(t)$ and of the corresponding adjoint state $\mathbf{p}(t)$ formed by the Lagrange multipliers, $\mathbf{p}(t) \equiv\left[p_{1}(t), \cdots, p_{N}(t)\right]$, fulfill Hamilton's equations

$$
\begin{equation*}
\dot{\mathbf{x}}=\frac{\partial H_{c}}{\partial \mathbf{p}}, \quad \dot{\mathbf{p}}=-\frac{\partial H_{c}}{\partial \mathbf{x}} \tag{3.116}
\end{equation*}
$$

associated to the control pseudo-Hamiltonian

$$
\begin{equation*}
H_{c}(\mathbf{p}(t), \mathbf{x}(t) ; \mathbf{u}(t))=\mathbf{p} \cdot \mathbf{f}(\mathbf{x}(t) ; \mathbf{u}(t))-p_{0} g(\mathbf{x}(t), \mathbf{u}(t)) \tag{3.117}
\end{equation*}
$$

where the constant $p_{0}>0$ can be chosen for convenience since it amounts to multiply the cost function by a constant. For almost all $t_{i} \leq t \leq t_{f}$ the function $H_{c}(\mathbf{p}(t), \mathbf{x}(t) ; \mathbf{u}(t))$ is maximum at certain controls $\mathbf{v}(t)=\mathbf{u}(t)$, for which one can write $H_{c}(\mathbf{p}(t), \mathbf{x}(t) ; \mathbf{v}(t))=$ const.

Our goal here is to minimize the energy:

$$
\begin{equation*}
J=\int_{t_{i}}^{t_{f}}\left(\Omega_{s}^{2}+\Omega_{p}^{2}\right) d t \tag{3.118}
\end{equation*}
$$

for which the control pseudo-Hamiltonian reads (with the standard choice $p_{0}=1 / 2$ )

$$
\begin{equation*}
H_{c}=\lambda^{\top}\left(\frac{1}{2} \Omega_{p} T_{1}+\frac{1}{2} \Omega_{s} T_{2}\right) \mathbf{x}-\frac{1}{2}\left(\Omega_{s}^{2}+\Omega_{p}^{2}\right) \tag{3.119}
\end{equation*}
$$

and the adjoint equation for the costate $\lambda:=\mathbf{p}^{\top}$ is determined from the Hamilton equations to be the same as the equation for $\mathbf{x}$ :

$$
\begin{equation*}
\dot{\lambda}=\frac{1}{2}\left(\Omega_{p} T_{1}+\Omega_{s} T_{2}\right) \lambda . \tag{3.120}
\end{equation*}
$$

We apply the maximum principle

$$
\begin{align*}
& \frac{\partial H_{c}}{\partial \Omega_{p}}=\frac{1}{2} \lambda^{\top} T_{1} \mathbf{x}-\Omega_{p}=0  \tag{3.121}\\
& \frac{\partial H_{c}}{\partial \Omega_{s}}=\frac{1}{2} \lambda^{\top} T_{2} \mathbf{x}-\Omega_{s}=0 \tag{3.122}
\end{align*}
$$

i.e.

$$
\begin{equation*}
\Omega_{p}=\frac{1}{2}\left(\lambda_{2} x_{1}-\lambda_{1} x_{2}\right), \quad \Omega_{s}=\frac{1}{2}\left(\lambda_{3} x_{2}-\lambda_{2} x_{3}\right) \tag{3.123}
\end{equation*}
$$

leading to

$$
\begin{equation*}
H_{c}=\frac{1}{2}\left(\Omega_{s}^{2}+\Omega_{p}^{2}\right) \tag{3.124}
\end{equation*}
$$

and

$$
\begin{align*}
\dot{\Omega}_{p} & =\omega \Omega_{s}  \tag{3.125a}\\
\dot{\Omega}_{s} & =-\omega \Omega_{p} \tag{3.125b}
\end{align*}
$$

noticing that

$$
\begin{equation*}
\omega \equiv \lambda^{\top} T_{3} \mathbf{x} / 4=\left(\lambda_{1} x_{3}-\lambda_{3} x_{1}\right) / 4 \tag{3.126}
\end{equation*}
$$

is a constant of motion. We can solve (3.125):

$$
\begin{equation*}
\Omega_{p}(t)=\Omega_{0} \cos (\omega t+\kappa), \quad \Omega_{s}(t)=-\Omega_{0} \sin (\omega t+\kappa) \tag{3.127}
\end{equation*}
$$

where we assume without loss of generality that $\Omega_{0}$ is positive. Using the boundary condition $x_{1}\left(t_{i} \equiv 0\right)=1$ and $x_{3}\left(t_{f} \equiv T\right)=1$, we get the following equations (i) at time $t=0$ :

$$
\begin{equation*}
\Omega_{p}(0)=\Omega_{0} \cos \kappa=\frac{1}{2} \lambda_{2}(0), \quad \Omega_{s}(0)=-\Omega_{0} \sin \kappa=0 \tag{3.128a}
\end{equation*}
$$

from which we can choose $\kappa=0$, and we get

$$
\begin{equation*}
\lambda_{2}(0)=2 \Omega_{0} \tag{3.128b}
\end{equation*}
$$

and (ii) at time $t=T$ :

$$
\begin{align*}
& \Omega_{p}(T)=\Omega_{0} \cos (\omega T)=0  \tag{3.128c}\\
& \Omega_{s}(T)=-\Omega_{0} \sin (\omega T)=-\frac{1}{2} \lambda_{2}(T) \tag{3.128d}
\end{align*}
$$

i.e.

$$
\begin{equation*}
\omega= \pm \frac{\pi}{2 T}, \quad \lambda_{2}(T)=\operatorname{sgn}(\omega) 2 \Omega_{0} \tag{3.129}
\end{equation*}
$$

We also have from Eq. (3.126)

$$
\begin{equation*}
\omega=-\frac{1}{4} \lambda_{3}(0)=\frac{1}{4} \lambda_{1}(T) \tag{3.130}
\end{equation*}
$$

And we have (conservation of the norm of the costate)

$$
\begin{equation*}
\lambda_{1}^{2}(0)+\lambda_{2}^{2}(0)+\lambda_{3}^{2}(0)=\lambda_{1}^{2}(T)+\lambda_{2}^{2}(T)+\lambda_{3}^{2}(T) \tag{3.131}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
\lambda_{1}^{2}(0)=\lambda_{3}^{2}(T) \tag{3.132}
\end{equation*}
$$

We can conclude that the optimal pulses, which satisfy the above boundary conditions, are of the form:

$$
\begin{equation*}
\Omega_{p}(t)=\Omega_{0} \cos \left(\frac{\pi t}{2 T}\right), \quad \Omega_{s}(t)=-\Omega_{0} \sin \left(\operatorname{sgn}(\omega) \frac{\pi t}{2 T}\right) \tag{3.133}
\end{equation*}
$$

They feature an intuitive sequence as it is well known, determined with the use of spherical coordinates which simplify the determination of optimal controls [20]. We show the details of the calculations below in Subsection 3.6.2. Here we would like to derive the optimal control problem keeping the original coordinates as it shows an explicit use of the shooting method. We have thus to solve both dynamical equations, (3.113) for the state and (3.120) for the costate, that satisfied the above boundary conditions: $x_{1}(0)=1$, $x_{3}(T)=1, \lambda_{2}(0)=\operatorname{sgn}(\omega) \lambda_{2}(T)=2 \Omega_{0}, \lambda_{1}(T)=-\lambda_{3}(0)=4 \omega= \pm \frac{2 \pi}{T}$. The unknown quantities are thus $\Omega_{0}$ and $\lambda_{1}(0)$, and also the sign of $\omega$. The shooting method consists in solving numerically both (3.113) and (3.120) with the initial conditions $x_{1}(0)=1$ and $\lambda_{2}(0)=2 \Omega_{0}, \lambda_{3}(0)=-4 \omega=\mp \frac{2 \pi}{T}$, for various $\Omega_{0}$ and various initial condition
$\lambda_{1}(0)$ (and testing both signs for $\omega$ ) and determine the final deviation

$$
\begin{equation*}
\varepsilon:=\left|x_{3}(T)-1\right|+\left|\lambda_{1}(T)+\lambda_{3}(0)\right|+\left|\lambda_{2}(T)-\operatorname{sgn}(\omega) \lambda_{2}(0)\right| \tag{3.134}
\end{equation*}
$$

which should be zero for the optimal solution.
Numerical simulations are shown in Figure 3.1 for $\omega=-\pi / 2 T$ (inducing $\lambda_{2}(T)=$ $-2 \Omega_{0}$ ) for which the deviation $\varepsilon$ can be found to be 0 , which occurs for

$$
\begin{equation*}
\Omega_{0}=\frac{\sqrt{3} \pi}{T}, \quad \lambda_{1}(0)=0 \tag{3.135}
\end{equation*}
$$

Obviously the $\sqrt{3} \pi$ factor is not fully exhibited from Figure 3.1, but only an approximation of it. We know the exact value of this factor from the calculation in spherical coordinates, see below. We can conclude that the optimal pulses read

$$
\begin{equation*}
\Omega_{p}(t)=\frac{\sqrt{3} \pi}{T} \cos \left(\frac{\pi t}{2 T}\right), \quad \Omega_{s}(t)=\frac{\sqrt{3} \pi}{T} \sin \left(\frac{\pi t}{2 T}\right) \tag{3.136}
\end{equation*}
$$

One can notice the intuitive sequence $\cos / \sin$, which is the opposite sequence with the same pulse shapes of the multi-mode driving derived in Section 2.4.

### 3.6.2 Spherical coordinates

We can use the spherical coordinates, which define the dynamical mode $\left|\phi_{0}(t)\right\rangle$, i.e. with $\theta\left(t_{i}=0\right)=0, \theta\left(t_{f}=T\right)=\pi / 2, \varphi(0)=\varphi(T)=0$. The equations of the dynamics

$$
\begin{align*}
\Omega_{p} / 2 & =\dot{\varphi} \cos \theta+\dot{\theta} \sin \theta \operatorname{cotan} \varphi  \tag{3.137a}\\
\Omega_{s} / 2 & =-\dot{\varphi} \sin \theta+\dot{\theta} \cos \theta \operatorname{cotan} \varphi \tag{3.137b}
\end{align*}
$$

where we denote $u_{p} \equiv \Omega_{p} / 2, u_{s} \equiv \Omega_{s} / 2$,

$$
\begin{align*}
\dot{\varphi} & =u_{p} \cos \theta-u_{s} \sin \theta  \tag{3.138a}\\
\dot{\theta} & =\left(u_{p} \sin \theta+u_{s} \cos \theta\right) \tan \varphi \tag{3.138b}
\end{align*}
$$

can be simplified as

$$
\begin{align*}
& \dot{\varphi}=v_{p}  \tag{3.139a}\\
& \dot{\theta}=-v_{s} \tan \varphi, \tag{3.139b}
\end{align*}
$$



Figure 3.1: Decimal logarithm of the deviation $\varepsilon$ for various $\Omega_{0} T$ and $\lambda_{1}(0) T$ for $\omega=$ $-\pi / 2 T$.
using a rotation on the control fields

$$
\left[\begin{array}{c}
v_{p}  \tag{3.140}\\
v_{s}
\end{array}\right]=\left[\begin{array}{cc}
\cos \theta & -\sin \theta \\
-\sin \theta & -\cos \theta
\end{array}\right]\left[\begin{array}{l}
u_{p} \\
u_{s}
\end{array}\right] .
$$

The minimization of the energy

$$
\begin{equation*}
\mathcal{E}=\int_{t_{i}}^{t_{f}}\left[u_{p}^{2}(t)+u_{s}^{2}(t)\right] d t=\int_{t_{i}}^{t_{f}}\left[v_{p}^{2}(t)+v_{s}^{2}(t)\right] d t \tag{3.141}
\end{equation*}
$$

leads to the control pseudo-Hamiltonian

$$
\begin{equation*}
H_{c}=\lambda_{\varphi} v_{p}-\lambda_{\theta} v_{s} \tan \varphi-\frac{1}{2}\left(v_{p}^{2}+v_{s}^{2}\right) \tag{3.142}
\end{equation*}
$$

with $\lambda=\left[\lambda_{\varphi}, \lambda_{\theta}\right]^{\top}$ the costate. The Hamilton equations lead to the equation of motion (3.139) and to

$$
\begin{equation*}
\dot{\lambda}_{\varphi}=\frac{v_{s}}{\cos ^{2} \varphi} \lambda_{\theta}, \quad \dot{\lambda}_{\theta}=0 \tag{3.143}
\end{equation*}
$$

This implies that $\lambda_{\theta}$ is a constant of motion. We apply the maximum principle:

$$
\begin{align*}
& \frac{\partial H_{c}}{\partial v_{p}}=\lambda_{\varphi}-v_{p}=0,  \tag{3.144a}\\
& \frac{\partial H_{c}}{\partial v_{s}}=-\lambda_{\theta} \tan \varphi-v_{s}=0, \tag{3.144b}
\end{align*}
$$

which leads to $\lambda_{\varphi}=v_{p}$ and $\lambda_{\theta} \tan \varphi=-v_{s}$, i.e.

$$
\begin{equation*}
H_{c}=\frac{1}{2}\left(v_{p}^{2}+v_{s}^{2}\right)=\frac{1}{2}\left(\lambda_{\varphi}^{2}+\lambda_{\theta}^{2} \tan ^{2} \varphi\right) . \tag{3.145}
\end{equation*}
$$

We determine the optimal trajectory by quadrature, using $H_{c}=C / 2=$ const., since $H_{c}$ features an effective autonomous system:

$$
\begin{align*}
\dot{\varphi} & = \pm \lambda_{\theta} \tan \varphi_{m} \sqrt{1-\frac{\tan ^{2} \varphi}{\tan ^{2} \varphi_{m}}},  \tag{3.146a}\\
\frac{d \varphi}{d \theta} & =\frac{\dot{\varphi}}{\dot{\theta}}= \pm \frac{\tan \varphi_{m}}{\tan ^{2} \varphi} \sqrt{1-\frac{\tan ^{2} \varphi}{\tan ^{2} \varphi_{m}}}  \tag{3.146b}\\
\tan \varphi_{m} & =\frac{\sqrt{C}}{\lambda_{\theta}} \tag{3.146c}
\end{align*}
$$

where we have assumed $\varphi \geq 0$ (which is satisfied for $u_{p}$ and $u_{s}$ both positive) and $\dot{\varphi}(t=$ $T / 2)=0$ by symmetry, leading to $\varphi(T / 2) \equiv \varphi_{m}$ maximum at $t=T / 2$ and positive (negative) branch of (3.146) for $t \in[0, T / 2], \varphi$ increasing from 0 to $\varphi_{m}(t \in[T / 2, T], \varphi$ decreasing from $\varphi_{m}$ to 0 ).

Taking into account the boundary conditions, we determine the solution of (3.146a):

$$
\begin{equation*}
\sin \varphi=\sin \varphi_{m} \sin (\pi t / T) \tag{3.147}
\end{equation*}
$$

with $\lambda_{\theta} T=\pi \cos \varphi_{m}$, and the one of (3.146b), $\theta_{+}\left(\varphi_{+}\right)$, for the positive branch of the geodesic (where $\varphi$ increases from 0 to $\varphi_{m}$, denoted $\varphi_{+}$):

$$
\begin{equation*}
\theta_{+}=\operatorname{atan}\left(\frac{\sin \varphi_{+}}{\sqrt{\tan ^{2} \varphi_{m}-\frac{\sin ^{2} \varphi_{+}}{\cos ^{2} \varphi_{m}}}}\right)-\cos \varphi_{m} \operatorname{asin}\left(\frac{\sin \varphi_{+}}{\sin \varphi_{m}}\right) . \tag{3.148}
\end{equation*}
$$

By imposing by symmetry that $\theta\left(\varphi_{m}\right)=\pi / 4$, we get

$$
\begin{equation*}
\varphi_{m}=\pi / 3, \tag{3.149}
\end{equation*}
$$

which leads to

$$
\begin{equation*}
\theta_{+}=\operatorname{atan}\left(\frac{\sin \varphi_{+}}{\sqrt{3-4 \sin ^{2} \varphi_{+}}}\right)-\frac{1}{2} \operatorname{asin}\left(\frac{2}{\sqrt{3}} \sin \varphi_{+}\right) \tag{3.150}
\end{equation*}
$$

This also leads to the time dependence of $\varphi$ :

$$
\begin{equation*}
\sin \varphi=\frac{\sqrt{3}}{2} \sin (\pi t / T) \tag{3.151}
\end{equation*}
$$

and consequently to the one of $\theta$ :

$$
\begin{equation*}
\theta=\operatorname{atan}\left(\frac{1}{2} \tan (\pi t / T)\right)-\frac{\pi t}{2 T} \tag{3.152}
\end{equation*}
$$

For the negative branch (i.e. $\varphi$ decreasing from $\varphi_{m}$ to 0 ), Eq. (3.151) still holds. The negative branch $\theta_{-}\left(\varphi_{-}\right)$of the geodesic reads:

$$
\begin{equation*}
\theta_{-}=\frac{\pi}{2}-\operatorname{atan}\left(\frac{\sin \varphi_{-}}{\sqrt{3-4 \sin ^{2} \varphi_{-}}}\right)+\frac{1}{2} \operatorname{asin}\left(\frac{2}{\sqrt{3}} \sin \varphi_{-}\right) \tag{3.153}
\end{equation*}
$$

We note that $\operatorname{asin}\left(\sin \varphi_{-} / \sin \varphi_{m}\right)=\pi-\pi t / T$ in this case since $\varphi_{m}$ is maximum. Equation (3.152) hold as well if we assume the definition

$$
\operatorname{atan}(x) \in \begin{cases}{[0, \pi / 2[ } & \text { for } x \geq 0  \tag{3.154}\\ ] \pi / 2, \pi[ & \text { otherwise }\end{cases}
$$

Inserting these relations into the definition of the pulses (3.137), we obtain for the optimal pulses with respect to the energy:

$$
\begin{equation*}
\Omega_{p}(t)=\frac{\sqrt{3} \pi}{T} \cos \left(\frac{\pi t}{2 T}\right), \quad \Omega_{s}(t)=\frac{\sqrt{3} \pi}{T} \sin \left(\frac{\pi t}{2 T}\right) \tag{3.155}
\end{equation*}
$$

One can remark that minimizing the $\operatorname{cost} \mathcal{E}$ is equivalent to minimizing the transfer-time for bounded controls $u_{p}^{2}+u_{s}^{2} \leq$ const.

If one considers the minimization of the effective pulse area defined as

$$
\begin{equation*}
\mathcal{A}=\int_{t_{i}}^{t_{f}} \sqrt{u_{p}^{2}(t)+u_{s}^{2}(t)} d t=\int_{t_{i}}^{t_{f}} \sqrt{v_{p}^{2}(t)+v_{s}^{2}(t)} d t \tag{3.156}
\end{equation*}
$$

one can remark that this quantity is time-reparametrization invariant as it leads to the same trajectory in the state space $(\theta, \varphi)$ for any time-reparametrization. Considering
indeed the time-reparametrization $s \equiv s(t)$ leads to the Schrödinger equation

$$
i \frac{\partial}{\partial s}|\widetilde{\psi}(s)\rangle=\left[\begin{array}{ccc}
0 & \widetilde{u}_{p}(s) & 0  \tag{3.157}\\
\widetilde{u}_{p}(s) & 0 & \widetilde{u}_{s}(s) \\
0 & \widetilde{u}_{s}(s) & 0
\end{array}\right]|\widetilde{\psi}(s)\rangle
$$

with $|\widetilde{\psi}(s)\rangle=|\psi(t)\rangle$ and $\widetilde{u}_{k}(s)=u_{k}(t) / \dot{s}, k=p, s$ and to the cost

$$
\begin{equation*}
\mathcal{A}=\int_{s_{i}}^{s_{f}} \frac{1}{\dot{s}} \sqrt{u_{p}^{2}(t)+u_{s}^{2}(t)} d s=\int_{s_{i}}^{s_{f}} \sqrt{\widetilde{u}_{p}^{2}(s)+\widetilde{u}_{s}^{2}(s)} d s \tag{3.158}
\end{equation*}
$$

with $s_{i}=s\left(t_{i}\right), s_{f}=s\left(t_{f}\right)$, which shows that we recover the original problem. Such cost leads to the same equation as before $\lambda_{\varphi}^{2}+\lambda_{\theta}^{2} \tan ^{2} \varphi=$ const. and thus to the same geodesic (3.150), (3.153) for any time-parametrization. The minimum pulse area for the Rabi frequency is thus $2 \mathcal{A}=\sqrt{3} \pi$. It can be viewed as the counterpart of the $\pi$-pulse transfer of a two-level system to the Raman three-state system. We can remark that the minimum is smaller than the one that would be induced by the successive application of the pump and Stokes pulses $(2 \mathcal{A}=2 \pi)$.

A convenient parametrization, symmetric around $t=T / 2$ (i.e. such that $\varphi(T-t)=$ $\varphi(t))$, such that $\varphi(T / 2)=\pi / 3$, is given by [20]:

$$
\varphi(t)=\left\{\begin{array}{cc}
0 & \text { if } t \leq 0 \text { or } t \geq T  \tag{3.159}\\
\frac{\pi}{3} \frac{e^{-\alpha t(t-T) / T^{2}}-1}{e^{\alpha / 4}-1} & \text { otherwise }
\end{array}\right.
$$

Using the (dimensionless) quantity $\alpha$ such that $\alpha \gg 1$ allows to start and finish the process with pulses close to zero. One can choose for instance $\alpha=36$ [20].

Another convenient parametrization, which is smooth, is given by

$$
\varphi(t)=\left\{\begin{array}{cc}
0 & \text { if } t \leq 0 \text { or } t \geq T  \tag{3.160}\\
\frac{\pi}{3} e^{4 \beta} \exp \left[\frac{\beta T^{2}}{t(t-T)}\right] & \text { otherwise }
\end{array}\right.
$$

The (dimensionless) quantity $\beta$ measures the width of the pulse. The value $\beta=2$ leads to pulses close to the preceding ones with $\alpha=36$.

We show in Figure 3.2 the time-dependence of $\varphi$ with both expressions given by Eqs. (3.159) and (3.160), respectively, and the corresponding $\theta$ determined from Eqs. (3.150) and (3.153), and in Figure 3.3 the corresponding control pulses. We notice that such pulses both start and end at 0 together featuring intuitive sequences.


Figure 3.2: Parameters $\varphi$ and $\theta$ (in units of $1 / T$ ) corresponding to Eq. (3.159) ( $\alpha=36$ ) (full lines) and Eq. (3.160) $(\beta=2)$ (dashed lines), respectively, as function of time.


Figure 3.3: Control pulses $u_{p}$ and $u_{s}$ (in units of $1 / T$ ) corresponding to Eq. (3.159) ( $\alpha=36$ ) (full lines) and Eq. (3.160) $(\beta=2)$ (dashed lines), respectively, as function of time.

### 3.7 Optimal Control by gradient method

In this section, we describe the numerical technique to implement the optimization by the gradient method, known as GRAPE (Gradient Ascent Pulse Engineering) [37].

We consider again the three-level $\Lambda$-system (2.31), in which the Hamiltonian is given by

$$
H(t)=\frac{1}{2}\left[\begin{array}{ccc}
0 & \Omega_{p}(t) & 0  \tag{3.161}\\
\Omega_{p}(t) & 0 & \Omega_{s}(t) \\
0 & \Omega_{s}(t) & 0
\end{array}\right]
$$

where $\Omega_{p}$ and $\Omega_{s}$ are the Rabi frequencies which are assumed positive and the detuning is zero. The time-dependent Schrödinger equation (TDSE) writes

$$
i \frac{\partial}{\partial t}\left[\begin{array}{l}
c_{1}  \tag{3.162}\\
c_{2} \\
c_{3}
\end{array}\right]=\frac{1}{2}\left[\begin{array}{ccc}
0 & \Omega_{p}(t) & 0 \\
\Omega_{p}(t) & 0 & \Omega_{s}(t) \\
0 & \Omega_{s}(t) & 0
\end{array}\right]\left[\begin{array}{l}
c_{1} \\
c_{2} \\
c_{3}
\end{array}\right]
$$

We expand below the gradient method on this problem for optimizing the controls with respect to the following cost:

$$
\begin{equation*}
J=\left[\int_{t_{i}}^{t_{f}} x_{2}(t)^{2} d t-\alpha\right]^{2}+\left[\int_{t_{i}}^{t_{f}}\left(u_{p}^{2}+u_{s}^{2}\right) d t\right]^{2}+g\left(\left|x\left(t_{f}\right)\right\rangle\right) \tag{3.163}
\end{equation*}
$$

where $u_{p} \equiv \Omega_{p} / 2, u_{s} \equiv \Omega_{s} / 2, \alpha$ is a given value, and $g\left(\left|x\left(t_{f}\right)\right\rangle\right)=1-\left\langle 3 \mid x\left(t_{f}\right)\right\rangle$, which attains its minimum when
(i) $\int_{t_{i}}^{t_{f}} x_{2}(t)^{2} d t \rightarrow \alpha$, meaning that the time area of the transient population in the excited state is given;
(ii) $\int_{t_{i}}^{t_{f}}\left(u_{p}^{2}+u_{s}^{2}\right) d t$ is minimum (i.e. minimization of the energy of the pulses);
(iii) $\left\langle 3 \mid x\left(t_{f}\right)\right\rangle \rightarrow 1$, which corresponds to an efficient population transfer.

Considering a given time area of the population in the excited state is motivated by controlling the loss of the system when losses appear from the excited state (through spontaneous emission), see details in Chapter 4.

As before, we denote $x_{1} \equiv c_{1}, x_{2} \equiv i c_{2}$ and $x_{3} \equiv-c_{3}$ so that all elements of the solution $|x(t)\rangle$ are real and positive, then Eq. (3.162) can be transformed into

$$
\begin{equation*}
\frac{d}{d t}|x(t)\rangle=A(t)|x(t)\rangle \tag{3.164}
\end{equation*}
$$

with the definitions

$$
A(t) \equiv\left[\begin{array}{ccc}
0 & -u_{p}(t) & 0  \tag{3.165}\\
u_{p}(t) & 0 & -u_{s}(t) \\
0 & u_{s}(t) & 0
\end{array}\right], \quad|x(t)\rangle \equiv\left[\begin{array}{c}
x_{1}(t) \\
x_{2}(t) \\
x_{3}(t)
\end{array}\right] .
$$

The method for minimizing $J$ is based on the use of its gradient with respect to the controls which iteratively lead to the minimum. Since the Schrödinger equation can be solved only numerically, the method needs a time discretization. The time discretization is denoted $t_{0}, t_{1}, \cdots, t_{N}$ with $N$ intervals, where the initial time $t_{i} \equiv t_{0}$, and the finial time $t_{f} \equiv t_{N}$, with $t_{n}=t_{0}+n d t, n=0, \cdots, N$.

The solution reads

$$
\begin{equation*}
|x(t+d t)\rangle \approx e^{A(t) d t}|x(t)\rangle, \tag{3.166}
\end{equation*}
$$

where we denote

$$
\begin{equation*}
A_{n} \equiv A\left(t_{n}\right), \quad|x\rangle_{n} \equiv\left|x\left(t_{n}\right)\right\rangle, \tag{3.167}
\end{equation*}
$$

and define the propagator

$$
\begin{equation*}
U_{n}=e^{A_{n} d t} . \tag{3.168}
\end{equation*}
$$

According to Eq. (3.166), we thus obtain

$$
\begin{align*}
\left|x\left(t_{1}\right)\right\rangle & \equiv|x\rangle_{1} \\
& \approx e^{A_{0} d t}|x\rangle_{0} \\
& \approx e^{A_{1} d t}|x\rangle_{0}  \tag{3.169}\\
& =U_{1}|x\rangle_{0},
\end{align*}
$$

where, $|x\rangle_{0}$ is the initial state $|1\rangle=[1,0,0]^{\top}$. The components of $\left|x\left(t_{1}\right)\right\rangle$ are denoted

$$
\left|x\left(t_{1}\right)\right\rangle \equiv|x\rangle_{1}=\left[\begin{array}{l}
x_{1}\left(t_{1}\right)  \tag{3.170}\\
x_{2}\left(t_{1}\right) \\
x_{3}\left(t_{1}\right)
\end{array}\right] \equiv\left[\begin{array}{l}
x_{1,1} \\
x_{2,1} \\
x_{3,1}
\end{array}\right],
$$

where the second index refers to time. The solution at time $t_{2}$ is

$$
\begin{align*}
\left|x\left(t_{2}\right)\right\rangle & \equiv|x\rangle_{2} \\
& \approx e^{A_{1} d t}|x\rangle_{1} \\
& \approx e^{A_{2} d t}|x\rangle_{1}  \tag{3.171}\\
& =U_{2} U_{1}|x\rangle_{0},
\end{align*}
$$

and at an arbitrary time $t_{m}$ :

$$
\begin{equation*}
|x\rangle_{m}=U_{m} U_{m-1} \cdots U_{1}|x\rangle_{0} \tag{3.172}
\end{equation*}
$$

The control parameters at time $t_{n}$ and at the iteration $(i)$ are denoted

$$
\begin{align*}
u_{p, n}^{(i)} & \equiv u_{p}^{(i)}\left(t_{n}\right)  \tag{3.173a}\\
u_{s, n}^{(i)} & \equiv u_{s}^{(i)}\left(t_{n}\right) \tag{3.173b}
\end{align*}
$$

We define a cost $J>0$ that we want to minimize at each iteration. $J$ depends on the controls at all times, i.e.

$$
\begin{equation*}
J \equiv J\left(\vec{u}_{p}, \vec{u}_{s}\right) \tag{3.174}
\end{equation*}
$$

where we define a time vector for the controls:

$$
\vec{u}_{p}=\left[\begin{array}{c}
u_{p, 1}  \tag{3.175}\\
u_{p, 2} \\
\vdots \\
u_{p, N}
\end{array}\right], \quad \vec{u}_{s}=\left[\begin{array}{c}
u_{s, 1} \\
u_{s, 2} \\
\vdots \\
u_{s, N}
\end{array}\right] .
$$

A simple illustration is shown in Figure 3.4.


Figure 3.4: Schematic representation of a control amplitude $u_{k}(t)$, consisting of $N$ discretization steps of duration $\Delta t=T / N$. During each step $n$, the control amplitude $u_{k, n}$ is constant. The vertical arrows represent the gradients of $J$, indicating how each amplitude $u_{k, n}$ should be modified in the next iteration to improve the cost $J$.

We define the rule of the iterations through the gradient of $J$ [52], which determines the pulse update:

$$
\begin{align*}
& \vec{u}_{p}^{(i+1)}=\vec{u}_{p}^{(i)}+\left.\varepsilon \frac{\partial J}{\partial \vec{u}_{p}}\right|_{\vec{u}_{p}^{(i)}},  \tag{3.176a}\\
& \vec{u}_{s}^{(i+1)}=\vec{u}_{s}^{(i)}+\left.\varepsilon \frac{\partial J}{\partial \vec{u}_{s}}\right|_{\vec{u}_{s}^{(i)}}, \tag{3.176b}
\end{align*}
$$

with a small quantity $\varepsilon<0$. We have then the property that $J$ decreases at each iteration as it is shown below: We determine $J$ at iteration $(i+1)$ from the one at iteration ( $i$ ):

$$
\begin{align*}
J^{(i+1)} & \equiv J\left(\vec{u}_{p}^{(i+1)}, \vec{u}_{s}^{(i+1)}\right) \\
& =J\left(\vec{u}_{p}^{(i)}+\left.\varepsilon \frac{\partial J}{\partial \vec{u}_{p}}\right|_{\vec{u}_{p}^{(i)}}, \vec{u}_{s}^{(i+1)}+\left.\varepsilon \frac{\partial J}{\partial \vec{u}_{s}}\right|_{\vec{u}_{s}^{(i)}}\right) \\
& =J\left(\vec{u}_{p}^{(i)}+\vec{\nabla}_{u_{p}} J, \vec{u}_{s}^{(i)}+\vec{\nabla}_{u_{s}} J\right), \tag{3.177}
\end{align*}
$$

using the property that, for a two variable function $\mathscr{F}(x, y), \mathscr{F}\left(x+h_{x}, y+h_{y}\right)=$ $\mathscr{F}(x, y)+\left[\begin{array}{ll}h_{x} & h_{y}\end{array}\right]\left[\begin{array}{c}\frac{\partial \mathscr{F}}{\partial x} \\ \frac{\partial \mathscr{F}}{\partial y}\end{array}\right]$ up to errors of order $h_{x}^{2}, h_{y}^{2}$, we calculate

$$
\begin{align*}
J^{(i+1)}=\underbrace{J\left(\vec{u}_{p}^{(i)}, \vec{u}_{s}^{(i)}\right)}_{J^{(i)}}+\varepsilon(\underbrace{\vec{\nabla}_{u_{p}} J \cdot \vec{\nabla}_{u_{p}} J}_{\sum_{n=1}^{N}\left(\frac{\partial J}{\partial u_{p, n}}\right)^{2}}+\underbrace{\vec{\nabla}_{u_{s}} J \cdot \vec{\nabla}_{u_{s}} J}_{\sum_{n=1}^{N}\left(\frac{\partial J}{\partial u_{s, n}}\right)^{2}}), \tag{3.178}
\end{align*}
$$

and, since $\varepsilon<0, \sum_{n=1}^{N}\left(\frac{\partial J}{\partial u_{p, n}}\right)^{2}>0$ and $\sum_{n=1}^{N}\left(\frac{\partial J}{\partial u_{s, n}}\right)^{2}>0$, we conclude

$$
\begin{equation*}
J^{(i+1)}<J^{(i)} . \tag{3.179}
\end{equation*}
$$

Based on the former analysis, $|x\rangle_{n}$ depends on $u_{p, n}, u_{s, n}$ and on $|x\rangle_{n-1},|x\rangle_{n+1}$ depends on $u_{p, n+1}, u_{s, n+1}$ and on $|x\rangle_{n}$, which means $|x\rangle_{n+1}$ also depends on $u_{p, n}, u_{s, n}$. Thus, $\left|x_{n}\right\rangle$ depends on the whole history of the controls until $t_{n}$ except at $t_{0}$ according to the definition (3.172) of the propagator: $u_{k, 1}, u_{k, 2}, \cdots, u_{k, n}(k=p$ or $s)$ and on the initial condition $|x\rangle_{0}$.

Using the trapezoidal rule, we obtain

$$
\begin{align*}
J \approx & {\left[\Delta t\left(x_{2,0}+x_{2, N}+2 \sum_{n=1}^{N-1} x_{2, n}\right)-\alpha\right]^{2} } \\
& +\Delta t\left(u_{p, 0}^{2}+u_{p, N}^{2}+2 \sum_{n=1}^{N-1} u_{p, n}^{2}+u_{s, 0}^{2}+u_{s, N}^{2}+2 \sum_{n=1}^{N-1} u_{s, n}^{2}\right)  \tag{3.180}\\
& +1-\left\langle 3 \mid x\left(t_{f}\right)\right\rangle,
\end{align*}
$$

where $\Delta t \equiv\left(t_{f}-t_{i}\right) / N$. In the following, we detemine the numerical derivatives of $J$ with respect to $u_{p, n}$ and $u_{s, n}$ in order to iterate the calculation of the controls in (3.176).

The derivative of $J$ with respect to $u_{k}\left(t_{n}\right)(k=p$ or $s)$ can be calculated as

$$
\frac{\partial J}{\partial u_{k, n}}=\sum_{m=n}^{N} \frac{\partial J}{\partial x_{2, m}} \cdot \frac{\partial x_{2, m}}{\partial u_{k, n}}+\left\{\begin{array}{ll}
4 \Delta t \cdot u_{k, n}, & n \neq 0, N  \tag{3.181}\\
2 \Delta t \cdot u_{k, n}, & n=0, N
\end{array}+\frac{\partial g}{\partial u_{k, n}}\right.
$$

with

$$
\begin{gather*}
\frac{\partial J}{\partial x_{2,0}}=\frac{\partial J}{\partial x_{2, N}}=2 \Delta t\left(\int_{t_{i}}^{t_{f}} x_{2}(t) d t-\alpha\right)  \tag{3.182a}\\
\frac{\partial J}{\partial x_{2, m_{(m \neq 0, N)}}}=4 \Delta t\left(\int_{t_{i}}^{t_{f}} x_{2}(t) d t-\alpha\right) \tag{3.182b}
\end{gather*}
$$

According to Eq. (3.172), we have for $1 \leq m \leq N$

$$
\begin{equation*}
x_{2, m}=\langle 2 \mid x\rangle_{m}=\langle 2| U_{m} U_{m-1} \cdots U_{1}|x\rangle_{0} \tag{3.183}
\end{equation*}
$$

which implies for $m>0$ and $n \leq m$ :

$$
\begin{equation*}
\frac{\partial x_{2, m}}{\partial u_{k, n}}=\langle 2| U_{m} U_{m-1} \cdots U_{n+1} \frac{\partial U_{n}}{\partial u_{k, n}} U_{n-1} \cdots U_{1}|x\rangle_{0} \tag{3.184}
\end{equation*}
$$

We have

$$
\begin{array}{ll}
\frac{\partial x_{2, m}}{\partial u_{k, 0}}=0, & \text { for } n=0 \\
\frac{\partial x_{2,0}}{\partial u_{k, n}}=0, & \text { for } m=0 \tag{3.186}
\end{array}
$$

The expansion of $U_{n}$ up to second order in $\Delta t$ based on Taylor's formula is as follows

$$
\begin{equation*}
U_{n}=e^{A_{n} \Delta t} \approx 1+A_{n} \Delta t+\frac{1}{2}\left(A_{n} \Delta t\right)^{2} \tag{3.187}
\end{equation*}
$$

From Eq. (3.165), we obtain

$$
\begin{gather*}
\frac{\partial A_{n}}{\partial u_{p, n}}=\left[\begin{array}{ccc}
0 & -1 & 0 \\
1 & 0 & 0 \\
0 & 0 & 0
\end{array}\right], \quad \frac{\partial A_{n}}{\partial u_{s, n}}=\left[\begin{array}{ccc}
0 & 0 & 0 \\
0 & 0 & -1 \\
0 & 1 & 0
\end{array}\right],  \tag{3.188}\\
A_{n}^{2}=\left[\begin{array}{ccc}
-u_{p, n}^{2} & 0 & u_{p, n} u_{s, n} \\
0 & -u_{p, n}^{2}-u_{s, n}^{2} & 0 \\
u_{p, n} u_{s, n} & 0 & -u_{s, n}^{2}
\end{array}\right]  \tag{3.189}\\
\frac{\partial A_{n}^{2}}{\partial u_{p, n}}=\left[\begin{array}{ccc}
-2 u_{p, n} & 0 & u_{s, n} \\
0 & -2 u_{p, n} & 0 \\
u_{s, n} & 0 & 0
\end{array}\right]  \tag{3.190a}\\
\frac{\partial A_{n}^{2}}{\partial u_{s, n}}=\left[\begin{array}{ccc}
0 & 0 & u_{p, n} \\
0 & -2 u_{s, n} & 0 \\
u_{p, n} & 0 & -2 u_{s, n}
\end{array}\right] \tag{3.190b}
\end{gather*}
$$

Combining equations from Eq. (3.187) to Eq. (3.190), we obtain for the derivative of $U n$ :

$$
\begin{align*}
\frac{\partial U_{n}}{\partial u_{p, n}} & =\frac{\partial A_{n}}{\partial u_{p, n}} \Delta t+\frac{(\Delta t)^{2}}{2} \frac{\partial A_{n}^{2}}{\partial u_{p, n}} \\
& =\Delta t\left(\frac{\partial A_{n}}{\partial u_{p, n}}+\frac{\Delta t}{2} \frac{\partial A_{n}^{2}}{\partial u_{p, n}}\right)  \tag{3.191}\\
& =\Delta t\left[\begin{array}{ccc}
-\Delta t u_{p, n} & -1 & \frac{\Delta t}{2} u_{s, n} \\
1 & -\Delta t u_{p, n} & 0 \\
\frac{\Delta t}{2} u_{s, n} & 0 & 0
\end{array}\right], \\
\frac{\partial U_{n}}{\partial u_{s, n}} & =\frac{\partial A_{n}}{\partial u_{s, n}} \Delta t+\frac{(\Delta t)^{2}}{2} \frac{\partial A_{n}^{2}}{\partial u_{s, n}} \\
& =\Delta t\left(\frac{\partial A_{n}}{\partial u_{s, n}}+\frac{\Delta t}{2} \frac{\partial A_{n}^{2}}{\partial u_{s, n}}\right)  \tag{3.192}\\
& =\Delta t\left[\begin{array}{ccc}
0 & 0 & \frac{\Delta t}{2} u_{p, n} \\
0 & -\Delta t u_{s, n} & -1 \\
\frac{\Delta t}{2} u_{p, n} & 1 & -\Delta t u_{s, n}
\end{array}\right] .
\end{align*}
$$

Taking Eq. (3.191) and Eq. (3.192) into Eq. (3.184), the derivative of $x_{2, m}$ with respect
to $u_{p, n}$ and $u_{s, n}$ can be obtained, respectively,

$$
\begin{align*}
& \frac{\partial x_{2, m}}{\partial u_{p, n}}=\Delta t \cdot{ }_{n+1}^{m}\langle\tilde{y}|\left[\begin{array}{ccc}
-\Delta t u_{p, n} & -1 & \frac{\Delta t}{2} u_{s, n} \\
1 & -\Delta t u_{p, n} & 0 \\
\frac{\Delta t}{2} u_{s, n} & 0 & 0
\end{array}\right]|x\rangle_{n-1},  \tag{3.193a}\\
& \frac{\partial x_{2, m}}{\partial u_{s, n}}=\Delta t \cdot{ }_{n+1}^{m}\langle\tilde{y}|\left[\begin{array}{ccc}
0 & 0 & \frac{\Delta t}{2} u_{p, n} \\
0 & -\Delta t u_{s, n} & -1 \\
\frac{\Delta t}{2} u_{p, n} & 1 & -\Delta t u_{s, n}
\end{array}\right]|x\rangle_{n-1}, \tag{3.193b}
\end{align*}
$$

where we denote

$$
\begin{equation*}
\underset{n+1}{m}\langle\tilde{y}|=\langle 2| U_{m} U_{m-1} \cdots U_{n+1} . \tag{3.194}
\end{equation*}
$$

Using explicitly the unitarity property of the Schrödinger equation

$$
i \frac{d}{d t}\left[\begin{array}{l}
x_{1}  \tag{3.195}\\
x_{2} \\
x_{3}
\end{array}\right]=i A(t)\left[\begin{array}{l}
x_{1} \\
x_{2} \\
x_{3}
\end{array}\right]
$$

denoting $C(t) \equiv i A(t)$, then we have $U_{C_{m}}=e^{-i C_{m} \Delta t}$, and $\left(U_{C_{m}}\right)^{\dagger}=e^{i C_{m} \Delta t}=e^{-A_{m} \Delta t}$. Therefore,

$$
\begin{equation*}
\left.\left({ }_{n+1}^{m}\langle\tilde{y}|\right)^{\dagger}=\langle 2| U_{m} U_{m-1} \cdots U_{n+1}\right)^{\dagger} \tag{3.196}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
|\tilde{y}\rangle_{n+1}^{m}=U_{n+1}^{\dagger} U_{n+2}^{\dagger} \cdots U_{m-1}^{\dagger} U_{m}^{\dagger}|2\rangle \tag{3.197}
\end{equation*}
$$

with $U_{m}=e^{A_{m} \Delta t}, U_{m}^{\dagger}=e^{-A_{m} \Delta t}$. Hence Eq. (3.197) can be written as

$$
\begin{equation*}
|\tilde{y}\rangle_{n+1}^{m}=e^{-A_{n+1} \Delta t} e^{-A_{n+2} \Delta t} \cdots e^{-A_{m-1} \Delta t} e^{-A_{m} \Delta t}|2\rangle \tag{3.198}
\end{equation*}
$$

Finally, we focus on the last part of the derivative (3.181) of the cost $J$, which only depends on the final state,

$$
\begin{equation*}
\frac{\partial g}{\partial u_{p, n}}=\frac{\partial g}{\partial\left|x\left(t_{f}\right)\right\rangle} \cdot \frac{\partial\left|x\left(t_{f}\right)\right\rangle}{\partial u_{p, n}} \tag{3.199}
\end{equation*}
$$

We calculate

$$
\begin{align*}
\frac{\partial g}{\partial\left|x\left(t_{f}\right)\right\rangle} & =\frac{\partial}{\partial\left|x\left(t_{f}\right)\right\rangle}\left(1-\left\langle 3 \mid x\left(t_{f}\right)\right\rangle\right) \\
& =\frac{\partial}{\partial\left|x\left(t_{f}\right)\right\rangle}\left(1-\left[\begin{array}{lll}
0 & 0 & 1
\end{array}\right]\left[\begin{array}{l}
x_{1}\left(t_{f}\right) \\
x_{2}\left(t_{f}\right) \\
x_{3}\left(t_{f}\right)
\end{array}\right]\right) \\
& =\left[\begin{array}{c}
0 \\
0 \\
-1
\end{array}\right] \equiv-|3\rangle \tag{3.200}
\end{align*}
$$

and

$$
\begin{equation*}
\frac{\partial\left|x\left(t_{f}\right)\right\rangle}{\partial u_{p, n}}=U_{N} \cdots U_{n+1} \frac{\partial U_{n}}{\partial u_{p, n}} U_{n-1} \cdots U_{1}|x\rangle_{0} \tag{3.201}
\end{equation*}
$$

Hence, Eq. (3.199) can be written as

$$
\begin{equation*}
\frac{\partial g}{\partial u_{p, n}}=-\langle 3| U_{N} \cdots U_{n+1} \frac{\partial U_{n}}{\partial u_{p, n}} U_{n-1} \cdots U_{1}|x\rangle_{0} \tag{3.202}
\end{equation*}
$$

We use the notation

$$
\begin{equation*}
|\tilde{z}\rangle_{n+1}^{N}=U_{n+1}^{\dagger} U_{n+2}^{\dagger} \cdots U_{N-1}^{\dagger} U_{N}^{\dagger}|3\rangle, \tag{3.203}
\end{equation*}
$$

with $U_{n}^{\dagger}=e^{-A_{n} d t}$, so the derivative of function $g$ with respect to $u_{p}\left(t_{n}\right)$ is

$$
\begin{equation*}
\frac{\partial g}{\partial u_{p, n}}={ }_{n+1}^{N}\langle\tilde{z}| \frac{\partial U_{n}}{\partial u_{p, n}}|x\rangle_{n-1} \tag{3.204}
\end{equation*}
$$

Combining this result with Eq. (3.191), we obtain

$$
\frac{\partial g}{\partial u_{p, n}}=-\Delta t \cdot{ }_{n+1}^{N}\langle\tilde{z}|\left[\begin{array}{ccc}
-\Delta t u_{p, n} & -1 & \frac{\Delta t}{2} u_{s, n}  \tag{3.205}\\
1 & -\Delta t u_{p, n} & 0 \\
\frac{\Delta t}{2} u_{s, n} & 0 & 0
\end{array}\right]|x\rangle_{n-1}
$$

Using the same method, we can obtain the derivative of function $g$ with respect to $u_{s}\left(t_{n}\right)$

$$
\frac{\partial g}{\partial u_{s, n}}=-\Delta t \cdot{ }_{n+1}^{N}\langle\tilde{z}|\left[\begin{array}{ccc}
0 & 0 & \frac{\Delta t}{2} u_{p, n}  \tag{3.206}\\
0 & -\Delta t u_{s, n} & -1 \\
\frac{\Delta t}{2} u_{p, n} & 1 & -\Delta t u_{s, n}
\end{array}\right]|x\rangle_{n-1}
$$

In conclusion, the GRAPE algorithm features the following properties:

- It uses forward- (3.183) and backward-propagations (3.197) and (3.203);
- Propagated states have to be stored;
- Pulse update (3.176) at time $n$ in the current iteration does not depend on other updated pulse values, thus all updates can in principle be calculated in parallel;
- The value of $\varepsilon$ has to be carefully chosen in order to reach the optimum. A too large value makes miss the optimum while a too small value makes the convergence very slow.

We will additionally see that

- As expected, it only finds a local minimum, and, in general, local minima are numerous;
- The obtained minimum will strongly depend on the initial guess of the parameters;
- The algorithm can evolve to a non-existing solution (from the constraint) that blocks it.


### 3.8 Trigonometric expansion for finite-time stimulated Raman exact passage (STIREP) by gradient method

In this section, we apply a systematic optimization procedure with the cost (3.163), but restrict the search through a finite trigonometric expansion of the angles taken from the dynamical mode (2.39a). The strong advantage of starting with the dynamical mode is that it ensures the exact population transfer when the appropriate boundaries of the angles are used. Incorporating ingredients of inverse engineering techniques in the optimization procedure allows its simplification and makes it more reliable.

Here we first focus on the following question: can we decrease the pulse energy obtained in Chapter 2 (pulses (2.49) for $k= \pm 1$ ) keeping the same transient time area on the excited state? We answer positively to this question investigating the search in a relatively small parameter space, in a systematic way. We also test this parameter space by trying to recover an approximation of the known optimal (in energy) solution.

### 3.8.1 Counterintuitive sequence as a starting point in a 8-parameter space

Considering (2.31), we assume as the starting point a finite-time counterintuitive STIREP process in the time interval $t \in[0, T]$, which implies $\Omega_{p}(t \leq 0)=0$ and $\Omega_{s}(t \geq T)=0$.

We use the convenient parametrization, corresponding to the dynamical mode (2.39a):

$$
|\psi\rangle=\left[\begin{array}{c}
\cos \phi \cos \vartheta  \tag{3.207}\\
i \sin \phi \\
\cos \phi \sin \vartheta
\end{array}\right],
$$

for which the field reads ( $u_{p} \equiv \Omega_{p} / 2 \leq 0$ and $u_{s} \equiv \Omega_{s} / 2 \geq 0$ ):

$$
\begin{align*}
& u_{p}=-\dot{\vartheta} \cot \phi \sin \vartheta-\dot{\phi} \cos \vartheta,  \tag{3.208a}\\
& u_{s}=\dot{\vartheta} \cot \phi \cos \vartheta-\dot{\phi} \sin \vartheta . \tag{3.208b}
\end{align*}
$$

Population transfer imposes the boundary conditions (with $\vartheta \geq 0$ and $\phi \geq 0$ )

$$
\begin{equation*}
0 \leftarrow \phi \rightarrow 0 \text { and } 0 \leftarrow \vartheta \rightarrow \pi / 2 . \tag{3.209}
\end{equation*}
$$

We make a trigonometric expansion of the angles, which is symmetric around time $T / 2$ :

$$
\begin{align*}
& \vartheta(t)=\frac{\pi}{4}+\sum_{m \geq 1} \vartheta_{2 m-1} \cos \frac{(2 m-1) \pi t}{T}, \quad \sum_{m \geq 1} \vartheta_{2 m-1}=-\frac{\pi}{4},  \tag{3.210a}\\
& \phi(t)=\sum_{n \geq 1} \phi_{2 n-1} \sin \frac{(2 n-1) \pi t}{T}, \tag{3.210b}
\end{align*}
$$

where $m, n$ are integers. The finite-time process imposes $u_{p}(0) \equiv \Omega_{p}(0) / 2=0$ and $u_{s}(T) \equiv \Omega_{s}(T) / 2=0$, i.e. the boundary conditions:

$$
\begin{align*}
& \dot{\phi}(0)=\dot{\phi}(T)=0,  \tag{3.211a}\\
& \dot{\vartheta}(0) \frac{\sin \vartheta(0)}{\sin \phi(0)}=\dot{\vartheta}(T) \frac{\cos \vartheta(T)}{\sin \phi(T)}=0 . \tag{3.211b}
\end{align*}
$$

This means

$$
\begin{equation*}
\sum_{n \geq 1}(2 n-1) \phi_{2 n-1}=0, \tag{3.212}
\end{equation*}
$$

and

$$
\begin{align*}
& \sum_{m \geq 1}(2 m-1)^{2} \vartheta_{2 m-1}=0  \tag{3.213a}\\
& \sum_{m \geq 1}(2 m-1)^{4} \vartheta_{2 m-1}=0 \tag{3.213b}
\end{align*}
$$

from

$$
\begin{align*}
\dot{\vartheta}(0) \frac{\sin \vartheta(0)}{\sin \phi(0)} & =-3 \frac{\pi}{T} \frac{\left[\sum_{m \geq 1}(2 m-1)^{2} \vartheta_{2 m-1}\right]^{2}}{\sum_{n \geq 1}(2 n-1)^{3} \phi_{2 n-1}}  \tag{3.214a}\\
\dot{\vartheta}(T) \frac{\cos \vartheta(T)}{\sin \phi(T)} & =-\frac{\pi}{T} \frac{\sum_{m \geq 1}(2 m-1)^{4} \vartheta_{2 m-1}}{\sum_{n \geq 1}(2 n-1)^{3} \phi_{2 n-1}} \tag{3.214b}
\end{align*}
$$

The cost (3.163) is only related to the energy and the transient population, since population transfer is automatically ensured with the boundary conditions (3.209). This shows here how one can incorporate ingredients of inverse engineering techniques in the optimization problem. The time area of the transient population in the excited state can be calculated as

$$
\begin{align*}
A & =\int_{0}^{T}|\langle 2 \mid \psi(t)\rangle|^{2} \\
& =\int_{0}^{T} \sin ^{2} \phi(t) d t \tag{3.215}
\end{align*}
$$

We choose a restricted parameter space with three parameters $\phi_{1}, \phi_{3}$ and $\phi_{5}$ for $\phi$, and five parameters $\vartheta_{1}, \vartheta_{3}, \vartheta_{5}$ and $\vartheta_{7}$ for $\vartheta$. The specific formulas of $\phi$ and $\vartheta$ reads:

$$
\begin{align*}
\vartheta(t) & =\frac{\pi}{4}+\vartheta_{1} \cos \frac{\pi t}{T}+\vartheta_{3} \cos \frac{3 \pi t}{T}+\vartheta_{5} \cos \frac{5 \pi t}{T}+\vartheta_{7} \cos \frac{7 \pi t}{T}+\vartheta_{9} \cos \frac{9 \pi t}{T}  \tag{3.216a}\\
\phi(t) & =\phi_{1} \sin \frac{\pi t}{T}+\phi_{3} \sin \frac{3 \pi t}{T}+\phi_{5} \sin \frac{5 \pi t}{T} \tag{3.216b}
\end{align*}
$$

The conditions Eq. (3.212) and Eq. (3.213) lead to the following relations among these parameters:

$$
\begin{equation*}
\phi_{5}=-\frac{1}{5} \phi_{1}-\frac{3}{5} \phi_{3}, \tag{3.217}
\end{equation*}
$$

and

$$
\begin{align*}
& \vartheta_{5}=-\frac{20}{7} \vartheta_{1}-\frac{15}{7} \vartheta_{3}-\frac{189}{256} \pi  \tag{3.218a}\\
& \vartheta_{7}=\frac{5}{2} \vartheta_{1}+\frac{3}{2} \vartheta_{3}+\frac{675}{1024} \pi  \tag{3.218b}\\
& \vartheta_{9}=-\frac{9}{14} \vartheta_{1}-\frac{5}{14} \vartheta_{3}-\frac{175}{1024} \pi \tag{3.218c}
\end{align*}
$$

This way we effectively need to compute the minimum over a space of only four independent coefficients (even though the number of parameters we controlled are eight). This allows one to apply a systematic procedure of optimization for such a relatively small parameter space. For a given value of the area on the excited state, the cost function attains its minimum when the minimum energy is reached.


Figure 3.5: Curve of the minimum energy with the variation of $\phi_{3}$ for a given value of $A=0.0879 T$ obtained from multi-mode driving.

Setting the time area of the transient population in the excited state $A=0.0879 T$ obtained from multi-mode driving, which gives $\mathcal{E}=\hbar \int\left(u_{p}^{2}+u_{s}^{2}\right) d t=\frac{15 \pi^{2}}{4} \frac{\hbar}{T} \simeq 37 \frac{\hbar}{T}$, we determine the minimum energy $23.6 \frac{\hbar}{T}$ for $\phi_{3}=-0.1850$ as shown in Figure 3.5 (and the values of other parameters are $\phi_{1}=0.3964, \phi_{5}=0.0317, \vartheta_{1}=-0.9560, \vartheta_{3}=0.2220$, $\vartheta_{5}=-0.0637, \vartheta_{7}=0.0139$ and $\vartheta_{9}=-0.0016$, respectively). The shapes of pulses and the corresponding populations as shown in Figure 3.6 and Figure 3.7. We see that the resulting pulses do not feature any more the intuitive sequence that was the starting point. They start (and end) together featuring a counterintuitive sequence near their peak amplitudes.

We conclude that we can notably decrease the value of the energy of the multi-mode solution (from $37 \frac{\hbar}{T}$ to $23.6 \frac{\hbar}{T}$ ) using a more complicated pulse shape. We, of course, do not pretend that this solution is optimal since it has been determined in a restricted parameter space.


Figure 3.6: Pump $u_{p}$ (solid blue line) and Stokes $u_{s}$ (solid red line) pulses with the parameters $\phi_{1}=0.3964, \phi_{3}=-0.1850, \vartheta_{1}=-0.9560$ and $\vartheta_{3}=0.2220$, that gives the smallest energy for a given $A=0.0879 T$ in the chosen 8 -parameter restricted space, compared to the pump (dashed cyan line) and Stokes (dashed magenta line) pulses derived by linear multi-mode driving.


Figure 3.7: Dynamics of the populations resulting from pulses (solid lines) of Figure 3.6. The transfer from state $|1\rangle$ to state $|3\rangle$ is exact due to the chosen boundaries (STIREP).

### 3.8.2 Intuitive sequence

We focus now on the intuitive sequence in this section, which imposes $u_{s}(t \leq 0)=0$ and $u_{p}(t \geq T)=0$, in order to optimize the cost in a restricted parameter space (similarly to the preceding subsection), imposing the time area on the excited state.

We know the global minimum energy for the intuitive process, $\mathcal{E}_{\text {glmin }}=3 \pi^{2} / 4 \approx$ $7.4022 \frac{\hbar}{T}$; global here means there is no condition on the area of the excited population, which is $A=0.3750 T$ in this case.

Starting from the trigonometric expansion by imposing $u_{s}(0)=0$ and $u_{p}(T)=0$, and combining with the boundary conditions in Eq. (3.209), we obtain

$$
\begin{equation*}
\sum_{m \geq 1}(2 m-1)^{4} \vartheta_{2 m-1}=0 \tag{3.219}
\end{equation*}
$$

from

$$
\begin{align*}
\frac{\dot{\vartheta}(0)}{\sin \phi(0)} & =-\frac{\pi}{T} \frac{\sum_{m \geq 1}(2 m-1)^{4} \vartheta_{2 m-1}}{\sum_{n \geq 1}(2 n-1)^{3} \phi_{2 n-1}}  \tag{3.220a}\\
\frac{\dot{\vartheta}(T)}{\sin \phi(T)} & =-\frac{\pi}{T} \frac{\sum_{m \geq 1}(2 m-1)^{4} \vartheta_{2 m-1}}{\sum_{n \geq 1}(2 n-1)^{3} \phi_{2 n-1}} \tag{3.220b}
\end{align*}
$$

By choosing two variable parameters for $\phi$ and two variable parameters for $\vartheta$, we know the expressions for two more parameters $\vartheta_{5}$ and $\vartheta_{7}$ :

$$
\begin{align*}
\vartheta_{5} & =-\frac{50}{37} \vartheta_{1}-\frac{145}{111} \vartheta_{3}-\frac{2401}{7104} \pi,  \tag{3.221a}\\
\vartheta_{7} & =\frac{13}{37} \vartheta_{1}+\frac{34}{111} \vartheta_{3}+\frac{625}{7104} \pi . \tag{3.221b}
\end{align*}
$$

Hence, the specific formulas of $\phi$ and $\vartheta$ reads:

$$
\begin{align*}
\vartheta(t) & =\frac{\pi}{4}+\vartheta_{1} \cos \frac{\pi t}{T}+\vartheta_{3} \cos \frac{3 \pi t}{T}+\vartheta_{5} \cos \frac{5 \pi t}{T}+\vartheta_{7} \cos \frac{7 \pi t}{T}  \tag{3.222a}\\
\phi(t) & =\phi_{1} \sin \frac{\pi t}{T}+\phi_{3} \sin \frac{3 \pi t}{T} \tag{3.222b}
\end{align*}
$$



Figure 3.8: Curve of the minimum energy with the variation of $\phi_{3}$ for a given $A=$ $0.3750 T$ ensuring the minimum energy in the 6 -parameter space.


Figure 3.9: Pump $u_{p}$ (solid blue line) and Stokes $u_{s}$ (solid red line) pulses with the parameters $\phi_{1}=0.9874, \phi_{3}=-0.0485, \vartheta_{1}=-0.9030, \vartheta_{3}=0.1330$ realizing the minimum energy in the 6 -parameter space, compared to the pump (dashed cyan line) and Stokes (dashed magenta line) pulses derived from (3.155).

Similarly to the analysis in Subsection 3.8.1, Figure 3.8 shows the variation of energy with respect to the parameter $\phi_{3}$ setting $A=0.3750 T$. The minimum energy in the 6 -parameter restricted space can be reached when $\phi_{3}=-0.0485$ (and the corresponding values of other parameters are $\phi_{1}=0.9874, \vartheta_{1}=-0.9030, \vartheta_{3}=0.1330, \vartheta_{5}=-0.0153$ and $\vartheta_{7}=-1.3860 \times 10^{-4}$, respectively). With such values of these parameters, the shapes of the pump and Stokes are shown in Figure 3.9, the corresponding populations being shown in Figure 3.10. For this particular value $A$, we should recover the optimal pulses (3.155), but since a restricted space is used, we only obtain an approximation of them (see Figure 3.9). We notice that the energy of these pulses is very slightly above the optimal one ( 7.42 vs 7.40 ). We next investigate other values of the transient population of the excited state as shown in Figure 3.11. The minimal energy decreases rapidly for increasing small values of $A$ and next seem to saturate around the value of the global optimum for larger $A$. In addition, we find that the value of the pump pulse at the initial time diminishes with decrease of the transient population of the excited state, and reaches zero when $A$ is reduced to some certain values.


Figure 3.10: Populations resulting from pulses (solid lines) of Figure 3.9.


Figure 3.11: Dependence of the minimum energy on the time area of the transient population in the excited state.

### 3.8.3 Gradient method

We finally investigate the gradient method, still in the 6 -parameter restricted space for simplication in a situation where the minimum is known (see previous subsection). We investigate two different initial conditions, one close to the minimum, and another far from the minimum. With the definition of the energy

$$
\begin{equation*}
\mathcal{E}=\int_{0}^{T}\left(u_{p}^{2}+u_{s}^{2}\right) d t, \tag{3.223}
\end{equation*}
$$

that we want to optimize during the iterations, we define the rule of iteration:

$$
\begin{align*}
& \vartheta_{2 m-1}^{(i+1)}=\vartheta_{2 m-1}^{(i)}+\left.\varepsilon \frac{\partial \mathcal{E}}{\partial \vartheta_{2 m-1}}\right|_{t},  \tag{3.224a}\\
& \phi_{2 n-1}^{(i+1)}=\phi_{2 n-1}^{(i)}+\left.\varepsilon \frac{\partial \mathcal{E}}{\partial \phi_{2 n-1}}\right|_{t}, \tag{3.224b}
\end{align*}
$$

with the choice $\varepsilon<0$ such that E decreases at each iteration.
The time discretization during the finite time is $t_{0}, t_{1}, \cdots, t_{N}$ with $N$ intervals, the initial time $t_{0} \equiv 0$, the time $t_{N} \equiv T$, and $\Delta t=T / N$.

Using trapezoidal rule, we can obtain

$$
\begin{equation*}
\mathcal{E} \approx \frac{\Delta t}{2}\left(u_{p t_{0}}^{2}+u_{p t_{N}}^{2}+2 \sum_{j=1}^{N-1} u_{p t_{j}}^{2}+u_{s t_{0}}^{2}+u_{s t_{N}}^{2}+2 \sum_{j=1}^{N-1} u_{s t_{j}}^{2}\right) . \tag{3.225}
\end{equation*}
$$

In order to calculate the iteration of the controls, we need the derivative of $\mathcal{E}$ with respect to $\theta_{2 m-1}$ and $\phi_{2 n-1}$, which can be calculated as

$$
\begin{align*}
& \left.\frac{\partial \mathcal{E}}{\partial \vartheta_{2 m-1}}\right|_{t}=2 \Delta t \sum_{j=1}^{N-1}\left(\frac{\partial \dot{\vartheta}_{t_{j}}}{\partial \vartheta_{2 m-1}} \dot{\vartheta}_{t_{j}} \cot ^{2} \phi_{t_{j}}\right),  \tag{3.226a}\\
& \left.\frac{\partial \mathcal{E}}{\partial \phi_{2 n-1}}\right|_{t}=2 \Delta t \sum_{j=1}^{N-1}\left(-\frac{\partial \phi_{t_{j}}}{\partial \phi_{2 n-1}} \dot{\vartheta}_{t_{j}}^{2} \frac{\cos \phi_{t_{j}}}{\sin ^{3} \phi_{t_{j}}}+\frac{\partial \dot{\phi}_{t_{j}}}{\partial \phi_{2 n-1}} \dot{\phi}_{t_{j}}\right), \tag{3.226b}
\end{align*}
$$

from

$$
\begin{align*}
\left.\frac{\partial \mathcal{E}}{\partial \vartheta_{2 m-1}}\right|_{t}= & \sum_{j=0}^{N}\left(\frac{\partial \mathcal{E}}{\partial u_{p_{t_{j}}}} \frac{\partial u_{p_{t_{j}}}}{\partial \vartheta_{t_{j}}} \frac{\partial \vartheta_{t_{j}}}{\partial \vartheta_{2 m-1}}+\frac{\partial \mathcal{E}}{\partial u_{p_{j}}} \frac{\partial u_{p_{t_{j}}}}{\partial \dot{\vartheta}_{t_{j}}} \frac{\partial \dot{\vartheta}_{t_{j}}}{\partial \vartheta_{2 m-1}}\right. \\
& \left.+\frac{\partial \mathcal{E}}{\partial u_{s t_{j}}} \frac{\partial u_{s t_{j}}}{\partial \vartheta_{t_{j}}} \frac{\partial \vartheta_{t_{j}}}{\partial \vartheta_{2 m-1}}+\frac{\partial \mathcal{E}}{\partial u_{s t_{j}}} \frac{\partial u_{s t_{j}}}{\partial \dot{\vartheta}_{t_{j}}} \frac{\partial \dot{\vartheta}_{t_{j}}}{\partial \vartheta_{2 m-1}}\right), \tag{3.227}
\end{align*}
$$

and

$$
\begin{align*}
\left.\frac{\partial \mathcal{E}}{\partial \phi_{2 n-1}}\right|_{t}= & \sum_{j=0}^{N}\left(\frac{\partial \mathcal{E}}{\partial u_{p_{t_{j}}}} \frac{\partial u_{p_{t_{j}}}}{\partial \phi_{t_{j}}} \frac{\partial \phi_{t_{j}}}{\partial \phi_{2 n-1}}+\frac{\partial \mathcal{E}}{\partial u_{p_{t_{j}}}} \frac{\partial u_{p_{t_{j}}}}{\partial \dot{\phi}_{t_{j}}} \frac{\partial \dot{\phi}_{t_{j}}}{\partial \phi_{2 n-1}}\right. \\
& \left.+\frac{\partial \mathcal{E}}{\partial u_{s t_{j}}} \frac{\partial u_{s t_{j}}}{\partial \phi_{t_{j}}} \frac{\partial \phi_{t_{j}}}{\partial \phi_{2 n-1}}+\frac{\partial \mathcal{E}}{\partial u_{s t_{j}}} \frac{\partial u_{s t_{j}}}{\partial \dot{\phi}_{t_{j}}} \frac{\partial \dot{\phi}_{t_{j}}}{\partial \phi_{2 n-1}}\right) \tag{3.228}
\end{align*}
$$

In combination with the parameters function Eq. (3.222), we know

$$
\begin{align*}
\left.\frac{\partial \mathcal{E}}{\partial \vartheta_{1}}\right|_{t} & =2 \Delta t \sum_{j=1}^{N-1}\left(\frac{\partial \dot{\vartheta}_{t_{j}}}{\partial \vartheta_{1}} \dot{\vartheta}_{t_{j}} \cot ^{2} \phi_{t_{j}}\right)  \tag{3.229a}\\
\left.\frac{\partial \mathcal{E}}{\partial \vartheta_{3}}\right|_{t} & =2 \Delta t \sum_{j=1}^{N-1}\left(\frac{\partial \dot{\vartheta}_{t_{j}}}{\partial \vartheta_{3}} \dot{\vartheta}_{t_{j}} \cot ^{2} \phi_{t_{j}}\right)  \tag{3.229b}\\
\left.\frac{\partial \mathcal{E}}{\partial \phi_{1}}\right|_{t} & =2 \Delta t \sum_{j=1}^{N-1}\left(-\frac{\partial \phi_{t_{j}}}{\partial \phi_{1}} \dot{\vartheta}_{t_{j}}^{2} \frac{\cos \phi_{t_{j}}}{\sin ^{3} \phi_{t_{j}}}+\frac{\partial \dot{\phi}_{t_{j}}}{\partial \phi_{1}} \dot{\phi}_{t_{j}}\right) \tag{3.229c}
\end{align*}
$$

with

$$
\begin{align*}
\frac{\partial \dot{\vartheta}_{t_{j}}}{\partial \vartheta_{1}} & =-\frac{\pi}{T}\left(\sin \frac{\pi t_{j}}{T}-\frac{250}{37} \sin \frac{5 \pi t_{j}}{T}+\frac{91}{37} \sin \frac{7 \pi t_{j}}{T}\right)  \tag{3.230a}\\
\frac{\partial \dot{\vartheta}_{t_{j}}}{\partial \vartheta_{3}} & =-\frac{\pi}{T}\left(3 \sin \frac{3 \pi t_{j}}{T}-\frac{725}{111} \sin \frac{5 \pi t_{j}}{T}+\frac{238}{111} \sin \frac{7 \pi t_{j}}{T}\right)  \tag{3.230b}\\
\frac{\partial \phi_{t_{j}}}{\partial \phi_{1}} & =\sin \frac{\pi t_{j}}{T}  \tag{3.230c}\\
\frac{\partial \dot{\phi}_{t_{j}}}{\partial \phi_{1}} & =\frac{\pi}{T} \cos \frac{\pi t_{j}}{T},  \tag{3.230d}\\
\dot{\vartheta}_{t_{j}} & =-\frac{\pi}{T}\left(\vartheta_{1} \sin \frac{\pi t_{j}}{T}+3 \vartheta_{3} \sin \frac{3 \pi t_{j}}{T}+5 \vartheta_{5} \sin \frac{5 \pi t_{j}}{T}+7 \vartheta_{7} \sin \frac{7 \pi t_{j}}{T}\right)  \tag{3.230e}\\
\dot{\phi}_{t_{j}} & =-\frac{\pi}{T}\left(\phi_{1} \cos \frac{\pi t_{j}}{T}+3 \phi_{3} \cos \frac{3 \pi t_{j}}{T}\right) \tag{3.230f}
\end{align*}
$$

and the modification of $\phi_{3}$ depends on both the given value of $A$ and the adapted value $\phi_{1}$ after the optimization.

We first choose the initial conditions of the iterations close to the absolute minimum energy (which is known from Subsection 3.8.2: $7.4239 \frac{\hbar}{T}$ for a given $A=0.3750 T$, see Figure 3.8). We show an example in Figure 3.12, in which the initial conditions are $\phi_{1}=1, \phi_{3}=0.2, \vartheta_{1}=-0.8$ and $\vartheta_{3}=0.1$. We obtain an optimal energy at the $101^{\text {th }}$ iteration as $7.6155 \frac{\hbar}{T}$. We conclude that the algorithm has been trapped in a
local minimum, i.e. located above the absolute minimum. Figure 3.12(b) shows the corresponding controls at the $101^{\text {th }}$ iteration. We find that both the values of the pump pulse and Stokes feature small but noticeable modifications with respect to the optimal ones.


Figure 3.12: (a) Energy optimization using the gradient method from initial conditions close to the known minimum energy (with the parameters $\phi_{1}=1, \phi_{3}=0.2, \vartheta_{1}=-0.8$, $\vartheta_{3}=0.1, \varepsilon=-0.00001$ ). (b) Pump $u_{p}$ (solid blue line) and Stokes $u_{s}$ (solid red line) pulses obtained from the optimization by the gradient method (at the $101^{\text {th }}$ iteration).

In order to recover the absolute optimum, we have tested various initial values of parameters. Starting with $\phi_{1}=1, \phi_{3}=0.2, \vartheta_{1}=-0.9030$ and $\vartheta_{3}=0.1330$ (i.e. taking the optimal values for $\vartheta_{1}, \vartheta_{3}$ in order to be closer to the optimal situation than before), we obtain the optimal energy at the $67^{\text {th }}$ iteration, as shown in Figure 3.13(a). The resulting pump and the Stokes pulses are depicted in Figure 3.13(b): we recover the pulses derived by the systematic optimization shown in Figure 3.9.

We next investigate the initial conditions far from the minimum. Nevertheless, for most of the examples, we could not get the solution through the gradient method. The simulation is interrupted due to the constraint of given value of $A$, which prevents to give existing solution in many cases.

In conclusion, through this study, even in a relatively small parameter space, we have highlighted the fact that the gradient method is a local optimization technique. In practice, it is thus difficult to distinguish whether the result obtained is a local optimal solution or a global optimal solution when the optimum is unknown. In addition, optimizing through the gradient method can lead to blockage due to absence of solutions satisfying the constraints for some initial values of the parameters.


Figure 3.13: (a) Energy optimization using the gradient method from initial conditions very close to the known minimum energy (with the parameters $\phi_{1}=1, \phi_{3}=0.2$, $\vartheta_{1}=-0.9030, \vartheta_{3}=0.1330, \varepsilon=-0.00001$ ). (b) Pump $u_{p}$ (solid blue line) and Stokes $u_{s}$ (solid red line) pulses obtained from the optimization by the gradient method (at the $67^{\text {th }}$ iteration).

## Optimal control for dissipative STIREP

## Contents

4.1 Definition of the dissipative driven stimulated Raman system ..... 70
4.2 Optimal control with respect to the energy of the controls con- strained by a given admissible loss: energy-optimal dissipative STIREP ..... 72
4.2.1 Construction of the pseudo-Hamiltonian ..... 72
4.2.2 Construction of the optimal trajectories from the Pontryagin max- imum principle ..... 76
4.2.3 Derivation of the pulses and the dynamics ..... 85
4.2.4 Comparison with standard STIRAP and parallel STIRAP ..... 93
4.2.5 Analogy with a pendulum ..... 97
4.3 Optimal control with respect to time constrained by a given admissible loss: time-optimal dissipative STIREP ..... 99
4.3.1 Construction of the pseudo-Hamiltonian ..... 99
4.3.2 Construction of the optimal trajectories from the Pontryagin max- imum principle ..... 102
4.3.3 Derivation of the pulses and the dynamics ..... 115
4.4 Discussion ..... 126

The object of this chapter is to apply optimal control techniques to optimize the population transfer in a Raman three-state system driven by two pulses (pump and Stokes pulses), i.e. minimizing the energy or time taking into account loss of the excited state. This is referred to as optimal dissipative STIREP. In the limit of low loss rate, a simplified and realistic model characterized the total loss consists in considering the time area of the transient population in the excited state $P_{2}(t)$. In this model, optimization will be then realized under the constraint of a given admissible loss (through a given time area $\left.A=\int P_{2}(t) d t\right)$. We show the following important results:

- when the given loss is relatively large, we obtain an intuitive pulse sequence pump/Stokes as optimal controls, similar to the optimal controls without constraints;
- when the given loss is low, we obtain as optimal controls an overlapping pulse sequence of specific shapes (see Eq. (4.101)) for energy optimization and a shaping featuring a transient counterintuitive pulse between fast intuitive sequences for time optimization with constrained pulse amplitudes (see Figures 4.22-4.27).
This is unexpected since it is widely believed that adiabatic passage using an approximate dark state (via counterintuitive pulse sequence Stokes/pump) is the most efficient one. This is not the case for the energy-minimization strategy: application of PMP shows that it is more efficient to operate fast via the excited state than to use long adiabatic passage accumulating lossy non-adiabatic population. The result shows that the pulses get shorter and more intense for lower loss. On the other hand, for the minimum-time strategy, we show that the dynamics operates relatively close to a dark state when the admissible loss is low.

This chapter is organized into four distinct sections. Section 4.1 is a brief overview of the analyzed system. In this section we define the three-level lossy quantum system and construct a simplified, though realistic, model for low dissipation rate. Section 4.2 is devoted to analyzing in detail the case of minimizing the energy of the controls constrained by an admissible loss. In Section 4.3, we present the time-optimal control of such system. Discussion and further comparison are given in Section 4.4.

### 4.1 Definition of the dissipative driven stimulated Raman system

We consider a three-level $\Lambda$-system, in which the Hamiltonian with rotating wave approximation (RWA) is given in the basis $\{|1\rangle,|2\rangle,|3\rangle\}$ by (in unit such that $\hbar=1$ )

$$
\hat{H}_{\Gamma}(t)=\left[\begin{array}{ccc}
0 & u_{p}(t) & 0  \tag{4.1}\\
u_{p}(t) & -i \Gamma / 2 & u_{s}(t) \\
0 & u_{s}(t) & 0
\end{array}\right],
$$

where $u_{p}$ and $u_{s}$ are the controls corresponding to half of the Rabi frequencies. Note that we have considered the pulses being exactly resonant since they produce the most efficient coupling. We consider a loss in the upper state via the dissipation rate $\Gamma$. Instead of analyzing such a lossy system, which is technically relatively difficult to treat in the optimal control frame (see Ref [53]), we consider alternatively the unlossy system $H \equiv H_{\Gamma=0}$ and the effect of the loss is taken into account at the second order perturbation theory from the knowledge of the state amplitude $c_{2}(t)$ of state 2 in absence
of dissipation:

$$
\begin{align*}
P_{\mathrm{loss}} & :=1-\left(\left|c_{\Gamma, 1}\right|^{2}+\left|c_{\Gamma, 2}\right|^{2}+\left|c_{\Gamma, 3}\right|^{2}\right)  \tag{4.2a}\\
& \approx \Gamma \int_{t_{i}}^{t_{f}} d t\left|c_{2}\right|^{2} \tag{4.2b}
\end{align*}
$$

where $t_{i}\left(t_{f}\right)$ is the initial (final) time, and the state amplitude of state $j$ is denoted $c_{\Gamma, j}$ in presence of the dissipation, and $c_{j}$ when $\Gamma=0$, respectively. This simplification is shown numerically to be valid for sufficiently low ratio $\Gamma / u_{\max }$ in Table 4.1.

| $\Gamma / u_{\max }$ | $1-\left(\left\|c_{\Gamma, 1}\right\|^{2}+\left\|c_{\Gamma, 2}\right\|^{2}+\left\|c_{\Gamma, 3}\right\|^{2}\right)$ | $\Gamma \int_{t_{i}}^{t_{f}} d t\left\|c_{2}\right\|^{2}$ |
| :---: | :---: | :---: |
| 0.1 | 0.0200 | 0.0224 |
| 0.08 | 0.0163 | 0.0179 |
| 0.06 | 0.0125 | 0.0134 |
| 0.04 | 0.0085 | 0.0090 |
| 0.02 | 0.0044 | 0.0045 |
| 0.01 | $2.2118 \times 10^{-3}$ | $2.2421 \times 10^{-3}$ |
| 0.008 | $1.7741 \times 10^{-3}$ | $1.7929 \times 10^{-3}$ |
| 0.006 | $1.3341 \times 10^{-3}$ | $1.3447 \times 10^{-3}$ |
| 0.005 | $1.1132 \times 10^{-3}$ | $1.1206 \times 10^{-3}$ |
| 0.004 | $8.9177 \times 10^{-4}$ | $8.9646 \times 10^{-4}$ |
| 0.002 | $4.4708 \times 10^{-4}$ | $4.4823 \times 10^{-4}$ |
| 0.001 | $2.2384 \times 10^{-4}$ | $2.2412 \times 10^{-4}$ |
| 0.0005 | $1.1200 \times 10^{-4}$ | $1.1206 \times 10^{-4}$ |

Table 4.1: Losses of the model (4.1) (4.2a) and the approximate model (4.4) (4.2b) for different and relatively low ratios $\Gamma / u_{\max }$ (obtained for example from standard STIRAP with Gaussian pulses with the peak of the pulses set to $\left.u_{\max }=6 / T\right)$. The approximate model provides reliable losses for the ratio $\Gamma / u_{\max }$ considered.

As before, denoting $x_{1} \equiv c_{1}, x_{2} \equiv i c_{2}$ and $x_{3} \equiv-c_{3}$ so that all the elements of the solution $x \equiv\left[x_{1}(t), x_{2}(t), x_{3}(t)\right]^{\top}$ are real when we consider the initial (real) state
$c_{1}\left(t_{i}\right)=1, c_{2}\left(t_{i}\right)=c_{3}\left(t_{i}\right)=0$, the TDSE becomes

$$
\begin{equation*}
\frac{d}{d t}|x\rangle=A(t)|x\rangle \tag{4.3}
\end{equation*}
$$

with

$$
A(t) \equiv\left[\begin{array}{ccc}
0 & -u_{p}(t) & 0  \tag{4.4}\\
u_{p}(t) & 0 & -u_{s}(t) \\
0 & u_{s}(t) & 0
\end{array}\right]
$$

satisfying $x_{1}^{2}+x_{2}^{2}+x_{3}^{2}=1$. The equation can be written as

$$
\begin{equation*}
\frac{d}{d t}|x\rangle=A(t)|x\rangle=\left(u_{p} T_{1}+u_{s} T_{2}\right)|x\rangle, \tag{4.5}
\end{equation*}
$$

with

$$
T_{1}=\left[\begin{array}{ccc}
0 & -1 & 0  \tag{4.6}\\
1 & 0 & 0 \\
0 & 0 & 0
\end{array}\right], \quad T_{2}=\left[\begin{array}{ccc}
0 & 0 & 0 \\
0 & 0 & -1 \\
0 & 1 & 0
\end{array}\right], \quad T_{3}=\left[\begin{array}{ccc}
0 & 0 & 1 \\
0 & 0 & 0 \\
-1 & 0 & 0
\end{array}\right],
$$

satisfying the commutation relations $\left[T_{1}, T_{2}\right]=T_{3},\left[T_{2}, T_{3}\right]=T_{1},\left[T_{3}, T_{1}\right]=T_{2}$.

### 4.2 Optimal control with respect to the energy of the controls constrained by a given admissible loss: energyoptimal dissipative STIREP

### 4.2.1 Construction of the pseudo-Hamiltonian

The goal of the control is to steer the system from $x_{1}\left(t_{i}\right)=1$ to $x_{3}\left(t_{f}\right)=1$ in a fxed time $t_{f}-t_{i}$ while minimizing the energy of the controls

$$
\begin{equation*}
J=\mathcal{E} \equiv \int_{t_{i}}^{t_{f}}\left(u_{p}^{2}+u_{s}^{2}\right) d t, \tag{4.7}
\end{equation*}
$$

under the loss constraint

$$
\begin{equation*}
\int_{t_{i}}^{t_{f}} d t x_{2}(t)^{2}=A \tag{4.8}
\end{equation*}
$$

with $A$ a given constant. This constraint characterizes the total admissible loss from the system, according to (4.2). $A$ has the unit of time, and will be expressed as a ratio of the total time of the process.

In order to take into account the constraint (4.8) in the PMP formalism, we augment
the dimension of the system with a new coordinate $y(t)$ such that

$$
\begin{equation*}
\dot{y}=x_{2}(t)^{2} \tag{4.9}
\end{equation*}
$$

of initial value $y\left(t_{i}\right)=0$ and final value $y\left(t_{f}\right)=\int_{t_{i}}^{t_{f}} d t x_{2}(t)^{2} \equiv A$. The constraint (4.8) reduces thus to a boundary problem on $y$.

The control pseudo-Hamiltonian of the augmented problem reads (with the standard choice $p_{0}=1 / 2$ )

$$
\begin{align*}
H_{c} & =\lambda^{\top}\left(u_{p} T_{1}+u_{s} T_{2}\right) x+\mu x_{2}^{2}-\frac{1}{2}\left(u_{p}^{2}+u_{s}^{2}\right) \\
& =-\lambda_{1} u_{p} x_{2}+\lambda_{2}\left(u_{p} x_{1}-u_{s} x_{3}\right)+\lambda_{3} u_{s} x_{2}+\mu x_{2}^{2}-\frac{1}{2}\left(u_{p}^{2}+u_{s}^{2}\right) \tag{4.10}
\end{align*}
$$

where the costate $\Lambda$, gathering the conjugate momenta of $x$ and $y$, has four components $\Lambda=\left[\lambda^{\top}, \mu\right]^{\top}$ with $\lambda=\left[\lambda_{1}, \lambda_{2}, \lambda_{3}\right]^{\top}$. The adjoint equations of the costate read

$$
\begin{align*}
& \dot{\lambda}^{\top}=-\frac{\partial H_{c}}{\partial x}  \tag{4.11a}\\
&=\left[\begin{array}{lll}
-\lambda_{2} u_{p}, & \lambda_{1} u_{p}-\lambda_{3} u_{s}-2 \mu x_{2}, & \lambda_{2} u_{s}
\end{array}\right]  \tag{4.11b}\\
& \dot{\mu}=-\frac{\partial H_{c}}{\partial y}
\end{align*}
$$

i.e.

$$
\begin{align*}
\dot{\lambda}_{1} & =-\lambda_{2} u_{p}  \tag{4.12a}\\
\dot{\lambda}_{2} & =\lambda_{1} u_{p}-\lambda_{3} u_{s}-2 \mu x_{2}  \tag{4.12b}\\
\dot{\lambda}_{3} & =\lambda_{2} u_{s}  \tag{4.12c}\\
\mu & =\text { const } \tag{4.12~d}
\end{align*}
$$

According to the maximum principle

$$
\begin{align*}
& \frac{\partial H_{c}}{\partial u_{p}}=-\lambda_{1} x_{2}+\lambda_{2} x_{1}-u_{p}=0  \tag{4.13a}\\
& \frac{\partial H_{c}}{\partial u_{s}}=-\lambda_{2} x_{3}+\lambda_{3} x_{2}-u_{s}=0 \tag{4.13b}
\end{align*}
$$

i.e., as for the unconstrained system (that is when (4.8) is not considered):

$$
\begin{align*}
& u_{p}=\lambda_{2} x_{1}-\lambda_{1} x_{2},  \tag{4.14a}\\
& u_{s}=\lambda_{3} x_{2}-\lambda_{2} x_{3}, \tag{4.14b}
\end{align*}
$$

which leads to

$$
\begin{equation*}
H_{c}=\frac{1}{2}\left(u_{p}^{2}+u_{s}^{2}\right)+\mu x_{2}^{2} \tag{4.15}
\end{equation*}
$$

and

$$
\begin{align*}
& \dot{u}_{p}=\omega u_{s}-2 \mu x_{1} x_{2}  \tag{4.16a}\\
& \dot{u}_{s}=-\omega u_{p}+2 \mu x_{2} x_{3}, \tag{4.16b}
\end{align*}
$$

noticing that

$$
\begin{equation*}
\omega \equiv \lambda^{\top} T_{3} x=\lambda_{1} x_{3}-\lambda_{3} x_{1} \tag{4.17}
\end{equation*}
$$

is a constant of motion (obtained from (4.12a), (4.12c) and (4.5)).

As seen in Chapter 3, it is convenient to use angle coordinates:

$$
\begin{align*}
& x_{1}=\cos \varphi \cos \theta  \tag{4.18a}\\
& x_{2}=\sin \varphi  \tag{4.18b}\\
& x_{3}=\cos \varphi \sin \theta \tag{4.18c}
\end{align*}
$$

with the initial condition $\varphi\left(t_{i}\right)=0, \theta\left(t_{i}\right)=0$. The equations of the dynamics (4.5)

$$
\begin{align*}
\dot{\varphi} & =u_{p} \cos \theta-u_{s} \sin \theta  \tag{4.19a}\\
\dot{\theta} & =\left(u_{p} \sin \theta+u_{s} \cos \theta\right) \tan \varphi \tag{4.19b}
\end{align*}
$$

can be simplified as

$$
\begin{align*}
\dot{\varphi} & =v_{p}  \tag{4.20a}\\
\dot{\theta} & =-v_{s} \tan \varphi \tag{4.20b}
\end{align*}
$$

after a rotation on the control fields

$$
\left[\begin{array}{l}
v_{p}  \tag{4.21}\\
v_{s}
\end{array}\right]=\left[\begin{array}{cc}
\cos \theta & -\sin \theta \\
-\sin \theta & -\cos \theta
\end{array}\right]\left[\begin{array}{l}
u_{p} \\
u_{s}
\end{array}\right]
$$

leading to an invariant cost on the new field variables since $u_{p}^{2}+u_{s}^{2}=v_{p}^{2}+v_{s}^{2}$. The minimization of the energy (4.7)

$$
\begin{equation*}
\mathcal{E}=\int_{t_{i}}^{t_{f}}\left[u_{p}^{2}(t)+u_{s}^{2}(t)\right] d t=\int_{t_{i}}^{t_{f}}\left[v_{p}^{2}(t)+v_{s}^{2}(t)\right] d t \tag{4.22}
\end{equation*}
$$

leads to the control pseudo-Hamiltonian

$$
\begin{equation*}
H_{c}=\lambda_{\varphi} v_{p}-\lambda_{\theta} v_{s} \tan \varphi+\mu \sin ^{2} \varphi-\frac{1}{2}\left(v_{p}^{2}+v_{s}^{2}\right) \tag{4.23}
\end{equation*}
$$

with $\Lambda=\left[\lambda_{\varphi}, \lambda_{\theta}, \mu\right]^{\top}$ the costate gathering the conjugate momenta of $\varphi, \theta$, and $y$, respectively. The Hamilton equations lead to the equation of motion (4.20) and to

$$
\begin{align*}
\dot{\lambda}_{\varphi} & =\frac{v_{s}}{\cos ^{2} \varphi} \lambda_{\theta}-\mu \sin (2 \varphi)  \tag{4.24}\\
\dot{\lambda}_{\theta} & =0  \tag{4.25}\\
\dot{\mu} & =0 \tag{4.26}
\end{align*}
$$

This implies that $\lambda_{\theta}$ and $\mu$ are constants of motion. We apply the maximum principle:

$$
\begin{align*}
& \frac{\partial H_{c}}{\partial v_{p}}=\lambda_{\varphi}-v_{p}=0  \tag{4.27a}\\
& \frac{\partial H_{c}}{\partial v_{s}}=-\lambda_{\theta} \tan \varphi-v_{s}=0 \tag{4.27b}
\end{align*}
$$

which gives $\lambda_{\varphi}=v_{p}$ and $\lambda_{\theta} \tan \varphi=-v_{s}$, leading to

$$
\begin{equation*}
H_{c}=\frac{1}{2}\left(\lambda_{\varphi}^{2}+\lambda_{\theta}^{2} \tan ^{2} \varphi\right)+\mu \sin ^{2} \varphi \tag{4.28}
\end{equation*}
$$

which features an effective autonomous system (i.e. explicitly time-independent since $H_{c}$ depends only on the dynamical variables and their conjugate momenta). The pseudoHamiltonian can be also rewritten as

$$
\begin{equation*}
H_{c}=\frac{1}{2}\left(v_{p}^{2}+v_{s}^{2}\right)+\mu \sin ^{2} \varphi \tag{4.29}
\end{equation*}
$$

The equations of motion finally read:

$$
\begin{align*}
\dot{\varphi} & =\lambda_{\varphi}  \tag{4.30a}\\
\dot{\theta} & =\lambda_{\theta} \tan ^{2} \varphi  \tag{4.30b}\\
\dot{y} & =\sin ^{2} \varphi  \tag{4.30c}\\
\dot{\lambda}_{\varphi} & =-\lambda_{\theta}^{2} \frac{\sin \varphi}{\cos ^{3} \varphi}-\mu \sin (2 \varphi)  \tag{4.30~d}\\
\dot{\lambda}_{\theta} & =\dot{\mu}=0 \tag{4.30e}
\end{align*}
$$

with the boundary conditions (for a complete population transfer from state $|1\rangle$ to state
$|3\rangle)$ :

$$
\begin{align*}
& \varphi\left(t_{i}\right)=0, \quad \theta\left(t_{i}\right)=0, \quad y\left(t_{i}\right)=0  \tag{4.31a}\\
& \varphi\left(t_{f}\right)=0, \quad \theta\left(t_{f}\right)=\pi / 2, \quad y\left(t_{f}\right)=A \tag{4.31b}
\end{align*}
$$

### 4.2.2 Construction of the optimal trajectories from the Pontryagin maximum principle

We present below the complete calculation of the optimal trajectories using PMP. The results will be regularly controlled with the unconstrained case $\mu=0$ developed in Chapter 3.

We assume a monotonically increasing $\theta$, implying $\lambda_{\theta}>0$. Since $H_{c}$ features an effective autonomous system, we can determine the optimal trajectory by quadrature using $H_{c}=h=$ const.

$$
\begin{align*}
\dot{\varphi} & =\lambda_{\varphi}= \pm \sqrt{2 h-\sin ^{2} \varphi\left(2 \mu+\frac{\lambda_{\theta}^{2}}{\cos ^{2} \varphi}\right)}  \tag{4.32a}\\
\frac{d \varphi}{d \theta} & =\frac{\dot{\varphi}}{\dot{\theta}}= \pm \frac{\sqrt{2 h-\sin ^{2} \varphi\left(2 \mu+\frac{\lambda_{\theta}^{2}}{\cos ^{2} \varphi}\right)}}{\lambda_{\theta} \tan ^{2} \varphi} \tag{4.32b}
\end{align*}
$$

We first integrate Eq. (4.32a) in order to find $\varphi\left(t ; h, \lambda_{\theta}, \mu\right)$, i.e. as a function of the three parameters $h, \lambda_{\theta}$, and $\mu$. These are determined to satisfy the boundary conditions for a given $A$.

We can assume $\varphi \geq 0$ (which is satisfied for $u_{p}$ and $u_{s}$ both positive), $t_{i} \equiv 0, t_{f} \equiv T$ and $\dot{\varphi}(t=T / 2)=0$ by symmetry, leading to $\varphi(T / 2) \equiv \varphi_{0}$ maximum at $t=T / 2$, and positive (negative) branch of (4.32) for $t \in[0, T / 2], \varphi$ increasing from 0 to $\varphi_{0}$ $\left(t \in[T / 2, T], \varphi\right.$ decreasing from $\varphi_{0}$ to 0$)$. This gives the relation:

$$
\begin{equation*}
2 h=\sin ^{2} \varphi_{0}\left(2 \mu+\frac{\lambda_{\theta}^{2}}{\cos ^{2} \varphi_{0}}\right) \tag{4.33}
\end{equation*}
$$

since $\lambda_{\theta}$ and $\mu$ are constant.
For the unconstrained system, i.e. $\mu=0$, this equation becomes

$$
\begin{equation*}
\sin ^{2} \varphi_{0}=\frac{2 h}{\lambda_{\theta}^{2}+2 h} \tag{4.34}
\end{equation*}
$$

i.e. we recover the condition for unconstrained $x_{2}(t)(3.146 \mathrm{c})$ :

$$
\begin{equation*}
\tan ^{2} \varphi_{0}=\frac{2 h}{\lambda_{\theta}^{2}} \tag{4.35}
\end{equation*}
$$

When $\mu \neq 0$, Eq. (4.33) can be rewritten as

$$
\begin{equation*}
\frac{\mu}{h} \sin ^{4} \varphi_{0}-\frac{2 h+2 \mu+\lambda_{\theta}^{2}}{2 h} \sin ^{2} \varphi_{0}+1=0 \tag{4.36}
\end{equation*}
$$

showing two relevant parameters

$$
\begin{equation*}
\alpha=\frac{\mu}{h}, \quad \beta=\frac{2 h+2 \mu+\lambda_{\theta}^{2}}{2 h}, \tag{4.37}
\end{equation*}
$$

as a function of which one can express two possible values of $\sin ^{2} \varphi_{0}$ :

$$
\begin{equation*}
\sin ^{2} \varphi_{0, \pm}=\frac{1}{2 \alpha}\left(\beta \pm \sqrt{\beta^{2}-4 \alpha}\right) \tag{4.38}
\end{equation*}
$$

which gives for $\alpha \rightarrow 0$ :

$$
\begin{equation*}
\sin ^{2} \varphi_{0, \pm}=\frac{1}{2 \alpha}\left(\beta \pm|\beta|\left(1-2 \alpha / \beta^{2}\right)\right) \tag{4.39}
\end{equation*}
$$

i.e. for $\beta>0$,

$$
\begin{equation*}
\sin ^{2} \varphi_{0, \pm}=\frac{1}{2 \alpha}(\beta \pm \beta \mp 2 \alpha / \beta), \tag{4.40}
\end{equation*}
$$

which leads to

$$
\begin{equation*}
\sin ^{2} \varphi_{0,-}=\frac{1}{\beta}, \tag{4.41}
\end{equation*}
$$

i.e. $\varphi_{0,-}=\pi / 3$ since we have $\beta=4 / 3$ for $\alpha=0$.

In general, this solution exists if $\beta^{2} \geq 4 \alpha$ and $\left(\beta \pm \sqrt{\beta^{2}-4 \alpha}\right) / \alpha>0$. If $\alpha>0$ and $\beta^{2}>4 \alpha$, there are two roots $\varphi_{0, \pm}$. If $\alpha<0$ and $\beta<0$, only $\varphi_{0,+}$ exists. If $\alpha<0$ and $\beta>0$, only $\varphi_{0,-}$ exists.

The differential equation (4.32a) can be rewritten as:

$$
\begin{align*}
\dot{\varphi} & = \pm \sqrt{2 h-\sin ^{2} \varphi\left(2 \mu+\frac{\lambda_{\theta}^{2}}{\cos ^{2} \varphi}\right)} \\
& = \pm \sqrt{2 h-\frac{\sin ^{2} \varphi}{\cos ^{2} \varphi}\left(2 \mu\left(1-\sin ^{2} \varphi\right)+\lambda_{\theta}^{2}\right)}  \tag{4.42a}\\
\dot{\varphi}|\cos \varphi| & = \pm \sqrt{2 h-\left(2 h+2 \mu+\lambda_{\theta}^{2}\right) \sin ^{2} \varphi+2 \mu \sin ^{4} \varphi} . \tag{4.42b}
\end{align*}
$$

Assuming $0 \leq \varphi \leq \pi / 2$, i.e. $\cos \varphi \geq 0$, we obtain:

$$
\begin{equation*}
\frac{1}{\sqrt{2 h}} \frac{d \sin \varphi}{d t}= \pm \sqrt{1-\beta \sin ^{2} \varphi+\alpha \sin ^{4} \varphi} \tag{4.43}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
\frac{d \sin \varphi}{\sqrt{1-\beta \sin ^{2} \varphi+\alpha \sin ^{4} \varphi}}= \pm \sqrt{2 h} d t \tag{4.44}
\end{equation*}
$$

assuming $h \geq 0$. Integrating the positive branch $\left(0 \leq t \leq T / 2,0 \leq \sin \varphi \leq \sin \varphi_{0}\right)$ leads to:

$$
\begin{equation*}
\int_{0}^{\sin \varphi} \frac{d s}{\sqrt{1-\beta s^{2}+\alpha s^{4}}}=\sqrt{2 h} t \tag{4.45}
\end{equation*}
$$

When $\mu=0$ (i.e. $\alpha=0$ ), we get

$$
\begin{equation*}
\frac{d \sin \varphi}{\sqrt{1-\sin ^{2} \varphi / \sin ^{2} \varphi_{0}}}= \pm \sqrt{2 h} d t \tag{4.46}
\end{equation*}
$$

which can be easily integrated; we obtain for the positive branch $(0 \leq t \leq T / 2)$ :

$$
\begin{equation*}
\sin ^{-1}\left(\sin \varphi / \sin \varphi_{0}\right) \sin \varphi_{0}=\sqrt{2 h} t \tag{4.47}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
\sin \varphi=\sin \varphi_{0} \sin \left(\frac{\sqrt{2 h}}{\sin \varphi_{0}} t\right)=\sin \varphi_{0} \sin \left(\frac{\lambda_{\theta}}{\cos \varphi_{0}} t\right) \tag{4.48}
\end{equation*}
$$

and at $t=T / 2$ :

$$
\begin{equation*}
\sin \varphi_{0}=\sin \varphi_{0} \sin \left(\frac{\lambda_{\theta} T}{2 \cos \varphi_{0}}\right) \tag{4.49}
\end{equation*}
$$

which is satisfied for

$$
\begin{equation*}
\lambda_{\theta} T=\pi \cos \varphi_{0} \tag{4.50}
\end{equation*}
$$

This gives (see (3.151))

$$
\begin{equation*}
\sin \varphi=\sin \varphi_{0} \sin (\pi t / T) \tag{4.51}
\end{equation*}
$$

When $\mu \neq 0$, the differential equation (4.45) involves an incomplete elliptic integral
of the first kind

$$
\begin{equation*}
F(\Phi \mid m)=\int_{0}^{\Phi} \frac{d \vartheta}{\sqrt{1-m \sin ^{2} \vartheta}} \tag{4.52}
\end{equation*}
$$

defined for $-\pi / 2<\Phi<\pi / 2$, and $m \sin ^{2} \Phi<1,0 \leq \vartheta \leq \Phi$, as follows: Defining $s=\kappa \sin \vartheta$, giving $d s=d \vartheta \kappa \cos \vartheta$ with $\cos \vartheta \geq 0$, i.e. $d \vartheta=d s /\left(\kappa \sqrt{1-(s / \kappa)^{2}}\right)$, we obtain:

$$
\begin{align*}
\kappa F(\Phi \mid m) & =\int_{0}^{\kappa \sin \Phi} \frac{d s}{\sqrt{\left(1-s^{2} / \kappa^{2}\right)\left(1-m s^{2} / \kappa^{2}\right)}} \\
& =\int_{0}^{\kappa \sin \Phi} \frac{d s}{\sqrt{1-(m+1) s^{2} / \kappa^{2}+m s^{4} / \kappa^{4}}} \tag{4.53}
\end{align*}
$$

By identification with (4.45), we have: $\beta=(m+1) / \kappa^{2}, \alpha=m / \kappa^{4}$, and $\sin \varphi=\kappa \sin \Phi$. We choose $\varphi=\varphi_{0}$ for $\Phi=\pi / 2$ (for which the elliptic integral of the first kind is complete), i.e.

$$
\begin{align*}
\kappa & =\sin \varphi_{0}  \tag{4.54a}\\
m & =\alpha \sin ^{4} \varphi_{0} \tag{4.54b}
\end{align*}
$$

and, for the positive branch (to be compared to (4.47) when $\mu=0$ ):

$$
\begin{equation*}
F\left(\sin ^{-1}\left(\sin \varphi / \sin \varphi_{0}\right) \mid m\right) \sin \varphi_{0}=\sqrt{2 h} t \tag{4.55}
\end{equation*}
$$

For $\mu=0$ (i.e. $\alpha=0$ ), we recover Eq. (4.47) from (4.55) for unconstrained $x_{2}(t)$ using the property

$$
\begin{equation*}
F(\Phi \mid 0)=\Phi \tag{4.56}
\end{equation*}
$$

Back to the general case, $\mu \neq 0$, at time $t=T / 2$, we have

$$
\begin{equation*}
F(\pi / 2 \mid m) \equiv K(m)=\frac{\sqrt{2 h} T}{2 \sin \varphi_{0}} \tag{4.57}
\end{equation*}
$$

where $K(m)$ is the complete elliptic integral of the first kind. This gives a condition on $\sqrt{2 h} / \sin \varphi_{0}$, which we insert into (4.55)

$$
\begin{equation*}
F\left(\sin ^{-1}\left(\sin \varphi / \sin \varphi_{0}\right) \mid m\right)=2 K(m) t / T \tag{4.58}
\end{equation*}
$$

The function $K(m)$ has the property to be real (and positive) when $m<1$, and $K(m) \rightarrow$ $\infty$ when $m \rightarrow 1$. The Jacobi elliptic functions allow one to invert the incomplete elliptic
integral of the first kind with respect to their first argument:

$$
\begin{align*}
& \operatorname{sn}(F(\Phi \mid m), m)=\sin \Phi  \tag{4.59a}\\
& \operatorname{cn}(F(\Phi \mid m), m)=\cos \Phi \tag{4.59b}
\end{align*}
$$

and (4.58) becomes (to be compared to (4.51) when $\mu=0$ ):

$$
\begin{equation*}
\sin \varphi=\sin \varphi_{0} \operatorname{sn}(2 K(m) t / T, m) \tag{4.60}
\end{equation*}
$$

We note that we have the property

$$
\begin{equation*}
\operatorname{sn}(K(m), m)=1 \tag{4.61}
\end{equation*}
$$

Equation (4.60) gives an explicit relation of $\sin \varphi$ as a function of time $0 \leq t \leq T / 2$ and of the two parameters $m$ and $\sin \varphi_{0}$. In order to take into account the boundary condition (4.30c), $\sin \varphi$ has to satisfy

$$
\begin{equation*}
\int_{0}^{T / 2} d t \sin ^{2} \varphi(t)=\frac{A}{2} \tag{4.62}
\end{equation*}
$$

We have next to integrate Eq. (4.32b) (still assuming $0 \leq \varphi \leq \pi / 2$, i.e. $\cos \varphi \geq 0$ ):

$$
\begin{equation*}
\frac{d \varphi}{d \theta}= \pm \sqrt{2 h} \frac{\sqrt{1-\beta \sin ^{2} \varphi+\alpha \sin ^{4} \varphi}}{\lambda_{\theta} \cos \varphi \tan ^{2} \varphi} \tag{4.63}
\end{equation*}
$$

where the sign $\pm$ is the same as the one of $\dot{\varphi}$, i.e.

$$
\begin{equation*}
\pm \frac{\sqrt{2 h}}{\lambda_{\theta}} d \theta=\frac{d \varphi}{\sqrt{1-\beta \sin ^{2} \varphi+\alpha \sin ^{4} \varphi}} \frac{\sin ^{2} \varphi}{\cos \varphi} \tag{4.64}
\end{equation*}
$$

that is, taking into account that $\theta=0$ when $\varphi=0$,

$$
\begin{equation*}
\theta= \pm \frac{\lambda_{\theta}}{\sqrt{2 h}} \int_{0}^{\varphi} \frac{d \phi}{\sqrt{1-\beta \sin ^{2} \phi+\alpha \sin ^{4} \phi}} \frac{\sin ^{2} \phi}{\cos \phi} \tag{4.65}
\end{equation*}
$$

with

$$
\begin{align*}
\frac{\lambda_{\theta}^{2}}{2 h} & =\beta-\alpha-1 \\
& =\frac{\sin ^{2} \varphi_{0}-m}{\sin ^{2} \varphi_{0} \tan ^{2} \varphi_{0}} \tag{4.66}
\end{align*}
$$

Since we have assumed $h>0$, we have

$$
\begin{equation*}
\sin ^{2} \varphi_{0} \geq m \tag{4.67}
\end{equation*}
$$

From the result:

$$
\begin{align*}
& \int_{0}^{\varphi} d \phi \frac{\sin ^{2} \phi}{\cos \phi \sqrt{\left(1-a \sin ^{2} \phi\right)\left(1-b \sin ^{2} \phi\right)}} \\
= & \frac{1}{\sqrt{a-b}}\left[F\left(\operatorname{asin}\left(\sqrt{1-a \sin ^{2} \varphi}\right) \left\lvert\, \frac{b}{b-a}\right.\right)-K\left(\frac{b}{b-a}\right)\right. \\
& \left.+\frac{a}{a-1} \Pi\left(\frac{1}{1-a} ;-\operatorname{asin}\left(\sqrt{1-a \sin ^{2} \varphi}\right) \left\lvert\, \frac{b}{b-a}\right.\right)+\frac{a}{a-1} \Pi\left(\left.\frac{1}{1-a} \right\rvert\, \frac{b}{b-a}\right)\right], \tag{4.68}
\end{align*}
$$

where $\Pi(n ; \Phi \mid m)$ is an incomplete elliptic integral of the third kind:

$$
\begin{equation*}
\Pi(n ; \Phi \mid m)=\int_{0}^{\Phi} \frac{d \phi}{\left(1-n \sin ^{2} \phi\right) \sqrt{1-m \sin ^{2} \phi}}, \tag{4.69}
\end{equation*}
$$

we identify:

$$
\begin{equation*}
\beta=a+b, \quad \alpha=a b, \tag{4.70}
\end{equation*}
$$

i.e.

$$
\begin{align*}
b^{2}-\beta b+\alpha & =0,  \tag{4.71a}\\
a & =\alpha / b, \tag{4.71b}
\end{align*}
$$

which gives

$$
\begin{align*}
b & =\frac{1}{2}\left(\beta \pm \sqrt{\beta^{2}-4 \alpha}\right) \\
& =\alpha \sin ^{2} \varphi_{0, \pm}, \tag{4.72}
\end{align*}
$$

and Eq. (4.65) becomes

$$
\begin{align*}
\theta= & \pm \frac{\sqrt{\sin ^{2} \varphi_{0}-m}}{\sqrt{1-m} \tan \varphi_{0}} \times\left[F\left(\left.\operatorname{asin} \sqrt{1-\frac{\sin ^{2} \varphi}{\sin ^{2} \varphi_{0}}} \right\rvert\, \frac{m}{m-1}\right)-K\left(\frac{m}{m-1}\right)\right. \\
& \left.+\frac{\Pi\left(-\tan ^{2} \varphi_{0} ; \left.-\operatorname{asin} \sqrt{1-\frac{\sin ^{2} \varphi}{\sin ^{2} \varphi_{0}}} \right\rvert\, \frac{m}{m-1}\right)}{\cos ^{2} \varphi_{0}}+\frac{\Pi\left(-\tan ^{2} \varphi_{0} \left\lvert\, \frac{m}{m-1}\right.\right)}{\cos ^{2} \varphi_{0}}\right] . \tag{4.73}
\end{align*}
$$

Imposing by symmetry that $\theta\left(\varphi_{0}\right)=\pi / 4$, the positive branch leads to

$$
\begin{equation*}
\frac{\pi}{4}=+\frac{\sqrt{\sin ^{2} \varphi_{0}-m}}{\sqrt{1-m} \tan \varphi_{0}} \times\left[-K\left(\frac{m}{m-1}\right)+\frac{\Pi\left(-\tan ^{2} \varphi_{0} \left\lvert\, \frac{m}{m-1}\right.\right)}{\cos ^{2} \varphi_{0}}\right] \tag{4.74}
\end{equation*}
$$

which gives an implicit relation between $\sin \varphi_{0}$ and $m$.
For $\mu=0$, i.e. $\alpha=0$ and $m=0$, we find $\varphi_{0}=\pi / 3$ as solution of (4.74) in accordance with (3.149), i.e. $\tan \varphi_{0} \equiv \sqrt{3}=\frac{\sqrt{2 h}}{\lambda_{\theta}}$ i.e. $a=0$ and $b=4 / 3$, and

$$
\begin{equation*}
\theta_{+}=\operatorname{atan}\left(\frac{\sin \varphi_{+}}{\sqrt{\tan ^{2} \varphi_{0}-\frac{\sin ^{2} \varphi_{+}}{\cos ^{2} \varphi_{0}}}}\right)-\cos \varphi_{0} \operatorname{asin}\left(\frac{\sin \varphi_{+}}{\sin \varphi_{0}}\right) \tag{4.75}
\end{equation*}
$$

Using

$$
\begin{equation*}
\operatorname{atan}(x)=\operatorname{asin}\left(\frac{x}{\sqrt{1+x^{2}}}\right) \tag{4.76}
\end{equation*}
$$

with

$$
\begin{equation*}
x=\frac{\sin \varphi_{+}}{\sqrt{\tan ^{2} \varphi_{0}-\frac{\sin ^{2} \varphi_{+}}{\cos ^{2} \varphi_{0}}}} \tag{4.77}
\end{equation*}
$$

we obtain

$$
\frac{x}{\sqrt{1+x^{2}}}=\frac{\frac{\sin \varphi_{+}}{\sqrt{\tan ^{2} \varphi_{0}-\frac{\sin ^{2} \varphi_{+}}{\cos ^{2} \varphi_{0}}}}}{\sqrt{1+\frac{\sin ^{2} \varphi_{+}}{\tan ^{2} \varphi_{0}-\frac{\sin ^{2} \varphi_{+}}{\cos ^{2} \varphi_{0}}}}}=\frac{\tan \varphi_{+}}{\tan \varphi_{0}}
$$

i.e.

$$
\begin{equation*}
\operatorname{atan}(x)=\operatorname{asin}\left(\frac{\tan \varphi_{+}}{\tan \varphi_{0}}\right) \tag{4.78}
\end{equation*}
$$

and $\theta_{+}$reads:

$$
\begin{equation*}
\theta_{+}=\operatorname{asin}\left(\frac{\tan \varphi_{+}}{\tan \varphi_{0}}\right)-\cos \varphi_{0} \operatorname{asin}\left(\frac{\sin \varphi_{+}}{\sin \varphi_{0}}\right) \tag{4.79}
\end{equation*}
$$

We have checked that this expression is equivalent to the one (Eq. (3.149) where $\varphi_{m} \equiv$ $\left.\varphi_{0}=\pi / 3\right)$ derived in Chapter 3.

The integral of the Jacobi elliptic function reads

$$
\begin{align*}
& \int d u \operatorname{sn}(u, m)^{2} \\
= & \frac{u \operatorname{dn}(u, m)-E(\operatorname{am}(u, m) \mid m) \sqrt{1-m \operatorname{sn}(u, m)^{2}}}{m \operatorname{dn}(u, m)} \tag{4.80}
\end{align*}
$$

where $E(\Phi \mid m)$ is an incomplete elliptic integral of the second kind, am $(u \mid m)$ is the
amplitude for Jacobi elliptic functions. Therefore, plugging Eq. (4.60) into Eq. (4.62), with the notation

$$
\begin{equation*}
u=2 K(m) t / T, \tag{4.81}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
d u=2 K(m) / T d t \tag{4.82}
\end{equation*}
$$

we obtain

$$
\begin{align*}
\frac{A}{2} & =\sin ^{2} \varphi_{0} \int_{0}^{T / 2} d t \operatorname{sn}(u, m)^{2} \\
& =\sin ^{2} \varphi_{0} \frac{T}{2 K(m)} \int_{0}^{K(m)} d u \operatorname{sn}(u, m)^{2} \\
& =\sin ^{2} \varphi_{0} \frac{T}{2 K(m)} \times\left.\frac{u \operatorname{dn}(u, m)-E(\operatorname{am}(u, m) \mid m) \sqrt{1-m \operatorname{sn}(u, m)^{2}}}{m \operatorname{dn}(u, m)}\right|_{0} ^{K(m)}  \tag{4.83}\\
& =\sin ^{2} \varphi_{0} \frac{T}{2 K(m)} \frac{K(m)-E(m)}{m} .
\end{align*}
$$

We determine the value of $A$ for the optimal unconstrained system, i.e. $m=0$ :

$$
\begin{equation*}
A\left(m=0, \varphi_{0}=\pi / 3\right)=\frac{3 T}{8} \tag{4.84}
\end{equation*}
$$

from the asymptotic result

$$
\begin{equation*}
\lim _{m \rightarrow 0} \frac{K(m)-E(m)}{m K(m)}=\frac{1}{2} . \tag{4.85}
\end{equation*}
$$

For a given value of $A$ (below $3 T / 8$, the one corresponding to the optimal unconstrained system), we aim at finding the optimal couple of the independent parameters $m$ and $\varphi_{0}$ simultaneously satisfying Eq. (4.74) and Eq. (4.83). The obtained data are shown in Table 4.2. The peak value $u_{\max }$ of each pulse and the pulse area $\mathcal{A}=\int_{0}^{T} d s \sqrt{u_{p}^{2}+u_{s}^{2}}$ are also provided in Table 4.2.

One can conclude that for decreasing $A, m$ decreases and $\varphi_{0}$ decreases to $\pi / 4$, both monotonically. This can be explained from Eq. (4.74), whose right-hand side goes to $\varphi_{0}$ when $m \rightarrow-\infty$. The minimum maximum value of $\varphi(t)$ is thus $\pi / 4$, asymptotically reached when $A \rightarrow 0$.

We note the lower energy (roughly twice) for the optimal pulse than for the multimode driving. We remark that the obtained minimal energy for $A=0.0879 T$ (area corresponding to the multi-mode driving $k=1$, $\mathcal{E}_{\min } \simeq 22.8 \frac{\hbar}{T}$, is relatively close to the one we obtained by systematic optimization in the restricted parameter space of 8-dimension in Section 3.8, $\mathcal{E}_{\text {min }}^{(8)} \simeq 23.6 \frac{\hbar}{T}$.

| $A$ (in units of $T)$ | $3 / 8$ | 0.35 | 0.3 | 0.25 | 0.2 |
| :---: | :---: | :---: | :---: | :---: | :---: |
| $m$ | 0 | -0.264 | -1.123 | -3.075 | -9.4 |
| $\varphi_{0}$ | $\pi / 3$ | 1.014 | 0.9502 | 0.8895 | 0.8364 |
| $\mathcal{E}$ (in units of $\hbar / T)$ | 7.40 | 7.44 | 7.79 | 8.64 | 10.28 |
| $u_{\max }($ in units of $1 / T)$ | 2.72 | 2.51 | 2.54 | 2.94 | 3.60 |
| $\mathcal{A}$ | 2.72 | 2.72 | 2.75 | 2.81 | 2.91 |


| $A$ (in units of $T)$ | 0.15 | 0.1 | 0.0879 | 0.08 | 0.07 |
| :---: | :---: | :---: | :---: | :---: | :---: |
| $m$ | -49.97 | -1379 | -5455 | -16770 | -100000 |
| $\varphi_{0}$ | 0.7995 | 0.7862 | 0.7856 | 0.7855 | 0.7854 |
| $\mathcal{E}$ (in units of $\hbar / T)$ | 13.40 | 20.00 | 22.75 | 25.00 | 28.57 |
| $u_{\max }($ in units of $1 / T)$ | 4.73 | 7.07 | 8.05 | 8.84 | 10.10 |
| $\mathcal{A}$ | 3.03 | 3.12 | 3.13 | 3.14 | 3.14 |


| $A$ (in units of $T$ ) | 0.06 | 0.05 | 0.04 | 0.03 |
| :---: | :---: | :---: | :---: | :---: |
| $m$ | -1082000 | -30320000 | $-4.5 \times 10^{9}$ | $-1.87 \times 10^{13}$ |
| $\varphi_{0}$ | 0.7854 | 0.7854 | 0.7854 | 0.7854 |
| $\mathcal{E}$ (in units of $\hbar / T)$ | 33.33 | 40.00 | 50.00 | 66.66 |
| $u_{\max }($ in units of $1 / T)$ | 11.79 | 14.14 | 17.68 | 23.57 |
| $\mathcal{A}$ | 3.14 | 3.14 | 3.14 | 3.14 |

Table 4.2: Optimal values of $m, \varphi_{0}$ and the corresponding energy $\mathcal{E}$ for various $A$. The value $A=0.0879 T$ corresponds to the multi-mode driving for $k=1$ (see Section 2.4). The peak value $u_{\max }$ and the pulse area $\mathcal{A}=\int_{0}^{T} d s \sqrt{u_{p}^{2}+u_{s}^{2}}$ are also given. The pulse area for the Rabi frequency is $2 \mathcal{A}$.

### 4.2.3 Derivation of the pulses and the dynamics

Equation (4.57) becomes

$$
\begin{equation*}
h=\frac{2 K(m)^{2} \sin ^{2} \varphi_{0}}{T^{2}} \tag{4.86}
\end{equation*}
$$

From Eq. (4.37), we derive

$$
\begin{align*}
\mu & =\alpha h,  \tag{4.87a}\\
\lambda_{\theta}^{2} & =2 h(\beta-\alpha-1), \tag{4.87b}
\end{align*}
$$

and both parameters $\alpha$ and $\beta$ can be expressed as functions of $m$ and $\varphi_{0}$ from Eq. (4.54):

$$
\begin{align*}
& \alpha=\frac{m}{\sin ^{4} \varphi_{0}},  \tag{4.88a}\\
& \beta=\frac{m+1}{\sin ^{2} \varphi_{0}} . \tag{4.88b}
\end{align*}
$$

From Eq. (4.21), the original controls can be written as

$$
\begin{align*}
& u_{p}=v_{p} \cos \theta-v_{s} \sin \theta,  \tag{4.89a}\\
& u_{s}=-v_{p} \sin \theta-v_{s} \cos \theta, \tag{4.89b}
\end{align*}
$$

where $\theta$ is given by Eq. (4.73), and

$$
\begin{align*}
& v_{p}=\lambda_{\varphi}= \pm \sqrt{2 h-\sin ^{2} \varphi\left(2 \mu+\frac{\lambda_{\theta}^{2}}{\cos ^{2} \varphi}\right)},  \tag{4.90a}\\
& v_{s}=-\lambda_{\theta} \tan \varphi, \tag{4.90b}
\end{align*}
$$

with $\varphi$ deduced from Eq. (4.60):

$$
\begin{equation*}
\varphi=\operatorname{asin}\left(\sin \varphi_{0} \operatorname{sn}(2 K(m) t / T, m)\right) \tag{4.91}
\end{equation*}
$$

From the values of $m$ and $\varphi_{0}$ shown in Table 4.2, one can derive the controls, the corresponding values of energy (shown in Table 4.2) and the resulting numerics of the populations.

The corresponding values of energy for the optimal couples of ( $m, \varphi_{0}$ ) according to Table 4.2 as a function of the time area of the transient population in the excited state $A$ are shown in Figure 4.1. For the unconstraint case, i.e. $m=0$, we recover the optimal
energy $\mathcal{E}=7.4022 \frac{\hbar}{T}$ and the corresponding optimal pulses (see Figure 4.2). Figure 4.1 shows a similar behavior as Figure 3.11: as the value of $A$ increases, the optimal energy decreases rapidly and next flattens out gradually towards the unconstraint situation, which gives the absolute smallest energy as expected.


Figure 4.1: Dependence of the optimal energy on the time area of the transient population in the excited state.

Figures 4.2 - 4.4 show the parameters $\varphi(t)$ and $\theta(t)$ and the dynamics for three typical values of $A$, respectively. Figure 4.5-4.8 compares controls for some values of $A$ of Table 4.2. We observe that the pump and Stokes controls get closer and coincide more and more for decreasing $A$.


Figure 4.2: Time-dependence of the parameters $\varphi$ (upper frame, solid cyan line) and $\theta$ (upper frame, solid magenta line) (in units of $1 / T$ ), the control pulses $u_{p}$ (middle frame, solid red line) and $u_{s}$ (middle frame, solid blue line) (in units of $1 / T$ ) and populations (lower frame) for the time area of the population in the excited state $A=$ $3 T / 8$ corresponding to unconstrained optimal pulses.


Figure 4.3: Same as Figure 4.2 but for $A=0.2 T$.


Figure 4.4: Same as Figure 4.2, but for $A=0.07 T$.


Figure 4.5: Controls for various values of $A=3 T / 8, A=0.3 T$ and $A=0.25 T$.


Figure 4.6: Same as Figure 4.5 but for $A=0.2 T, A=0.15 T$ and $A=0.1 T$.
4.2. Optimal control with respect to the energy of the controls constrained by a given admissible loss: energy-optimal dissipative STIREP


Figure 4.7: Controls for $A=0.08 T$. The pump and Stokes controls appear as overlapping pulses at the scale of the figure for $A \lesssim 0.08 T$.


Figure 4.8: Controls for various values of $A<0.08 T$.

In the limit $m \rightarrow-\infty, \varphi_{0}=\pi / 4$, we have

$$
\begin{align*}
2 \mu+\lambda_{\theta}^{2} & =2 \alpha h+2 h(\beta-\alpha-1)=2 h(2 m+1) \rightarrow 4 h m \rightarrow-\infty  \tag{4.92}\\
\mu & =\frac{4 m K(m)^{2}}{T^{2}} \rightarrow-\infty  \tag{4.93}\\
\lambda_{\theta}^{2} & =h(-4 m+2) \rightarrow-4 \frac{m K(m)^{2}}{T^{2}} \rightarrow+\infty \tag{4.94}
\end{align*}
$$

i.e.

$$
\begin{align*}
& v_{p}= \pm \sqrt{-4 m h} \sqrt{\cos 2 \varphi} \tan \varphi  \tag{4.95a}\\
& v_{s}=-\sqrt{-4 m h} \tan \varphi \tag{4.95b}
\end{align*}
$$

with

$$
\begin{equation*}
\sin \varphi=\frac{1}{\sqrt{2}} \operatorname{sn}(2 K(m) t / T, m) \tag{4.96}
\end{equation*}
$$

Eq. (4.73) becomes for the positive branch

$$
\begin{equation*}
\theta_{+}=-\operatorname{atan} \sqrt{1-\frac{\sin ^{2} \varphi_{+}}{\sin ^{2} \varphi_{0}}}+\frac{\pi}{4} \tag{4.97}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
-\tan \left(\theta_{+}-\frac{\pi}{4}\right)=\frac{1-\tan \theta_{+}}{1+\tan \theta_{+}}=\sqrt{1-\frac{\sin ^{2} \varphi_{+}}{\sin ^{2} \varphi_{0}}} \tag{4.98}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
\sin \varphi_{+}=\sin \varphi_{0} \frac{2 \sqrt{\tan \theta_{+}}}{1+\tan \theta_{+}} \tag{4.99}
\end{equation*}
$$

Comparing with (4.91), we get

$$
\begin{equation*}
\frac{2 \sqrt{\tan \theta_{+}}}{1+\tan \theta_{+}}=\operatorname{sn}(2 K(m) t / T, m) \tag{4.100}
\end{equation*}
$$

We finally find that, in the limit of a small area $A$, i.e. $A \lesssim 0.08 T$, corresponding to a large negative $m$ and $\varphi_{0}=\pi / 4$ :

$$
\begin{equation*}
u_{p}(t)=u_{s}(t)=\frac{\sqrt{-2 m} K(m)}{T} \operatorname{sn}\left(\frac{2 K(m) t}{T}, m\right) \tag{4.101}
\end{equation*}
$$

with $m$ solution of Eq. (4.83)

$$
\begin{equation*}
A=\frac{T}{2} \frac{K(m)-E(m)}{m K(m)}, \tag{4.102}
\end{equation*}
$$

which is well approximated by

$$
\begin{equation*}
m=-\frac{1}{2^{4}} e^{T / A} . \tag{4.103}
\end{equation*}
$$

In this limit, the peak of the pulses is well approximated by

$$
\begin{align*}
\max \left(u_{p}(t)\right)=\max \left(u_{s}(t)\right) & =\frac{\sqrt{-2 m} K(m)}{T} \\
& \approx \frac{\ln (16|m|)}{\sqrt{2} T} \\
& =\frac{1}{\sqrt{2} A}, \tag{4.104}
\end{align*}
$$

and the area of the pulses is

$$
\begin{equation*}
\int_{0}^{T} u_{p}(t) d t=\int_{0}^{T} u_{s}(t) d t=\frac{\pi}{\sqrt{2}} \tag{4.105}
\end{equation*}
$$

and the Rabi frequency pulse area

$$
\begin{equation*}
2 \int_{0}^{T} \sqrt{u_{p}^{2}(t)+u_{s}^{2}(t)} d t=2 \pi \tag{4.106}
\end{equation*}
$$

which corresponds to twice the minimum Rabi frequency pulse area.

### 4.2.4 Comparison with standard STIRAP and parallel STIRAP

We show as an example the dynamics in Figure 4.9(a) for the optimal STIREP with a low time area $A \simeq 0.036 T$. The dynamics and the energy of the derived pulses are compared to the ones of the standard STIRAP with Gaussian pulses in a situation giving the same time area $A$ (see Figure 4.9(b)). We obtain the energy $\mathcal{E} \simeq 90 \frac{\hbar}{T}$ for STIRAP, almost twice larger than energy-optimal STIREP $\mathcal{E} \simeq 56 \frac{\hbar}{T}$. Comparing the two figures in Figure 4.9, we conclude that the optimal pulses we derived feature a shorter duration, compared to the long adiabatic process of STIRAP, and are more intense with the peak $u_{\max } \simeq 19.6 / T$ three times larger than those of the standard STIRAP $u_{\max } \simeq 6 / T$.


Figure 4.9: Comparison of the dynamics between (a) the energy-optimal STIREP and (b) the standard STIRAP featuring the same time area in the excited state $A \simeq 0.036 T$.

We next make a comparison with respect to Stimulated Raman parallel adiabatic passage (parallel STIRAP) with coinciding pulses. For such a process, we have to consider quasi-resonant pulses [54, 55]:

$$
\mathrm{H}=\left[\begin{array}{ccc}
-\delta / 2 & u_{P} & 0  \tag{4.107}\\
u_{P} & \Delta & u_{S} \\
0 & u_{S} & \delta / 2
\end{array}\right]
$$

with $u_{p} \equiv \Omega_{P} / 2, u_{s} \equiv \Omega_{S} / 2$, and $\delta$ the two-photon detuning: $\delta=\omega_{3}-\omega_{1}-\omega_{P}+\omega_{S}, \Delta$ connected to the one-photon detuning (with respect to the pump) $\Delta_{P}=\omega_{2}-\omega_{1}-\omega_{P}$ as $\Delta=\Delta_{P}-\delta / 2 . \hbar \omega_{j}, j=1,2,3$ are the energies of the corresponding state $|j\rangle$ and $\omega_{P}, \omega_{S}$ the frequencies of the pump and Stokes fields, respectively. Parallel STIRAP requires an adiabatic passage process such that the eigenvalues stay parallel at each time, implying minimization of non-adiabatic coupling, which is a way to optimize the adiabatic passage [56]. Denote $\omega_{-, 0,+}$ the three eigenvalues which satisfy $\omega_{-}<\omega_{0}<\omega_{+}$ and $\left|\psi_{-, 0,+}\right\rangle$ the corresponding eigenstates such that $|1\rangle^{t \rightarrow-\infty} \stackrel{ }{\longleftrightarrow}\left|\psi_{0}\right\rangle^{t \rightarrow+\infty}|3\rangle$. The eigenvalues, on which we impose parallelism: $\omega_{+}-\omega_{0}=\omega_{0}-\omega_{-}=\Omega_{0} / 2$, are thus of the form

$$
\begin{equation*}
\omega_{0}=\frac{1}{3} \Delta, \quad \omega_{ \pm}=\frac{1}{3} \Delta \pm \frac{1}{2} \Omega_{0} \tag{4.108}
\end{equation*}
$$

with

$$
\begin{equation*}
\Omega_{0}=\sqrt{\Omega_{P}^{2}+\Omega_{S}^{2}+\delta^{2}+\frac{4}{3} \Delta^{2}} \tag{4.109}
\end{equation*}
$$

and the condition

$$
\begin{equation*}
0=\frac{\delta}{2}\left(\Omega_{S}^{2}-\Omega_{P}^{2}\right)+\frac{\Delta}{3}\left(\Omega_{P}^{2}+\Omega_{S}^{2}\right)+\frac{\Delta}{27}\left(8 \Delta^{2}-18 \delta^{2}\right) . \tag{4.110}
\end{equation*}
$$

We choose the initial and final connections, when $\Omega_{P}=\Omega_{S}=0, \omega_{+}(-\infty)=\Omega_{0} / 4$, $\omega_{-}(-\infty)=-3 \Omega_{0} / 4, \omega_{+}(+\infty)=3 \Omega_{0} / 4, \omega_{-}(+\infty)=-\Omega_{0} / 4$ corresponding to the final and initial conditions: $0 \stackrel{t \rightarrow-\infty}{\leftrightarrows} \Omega_{P, S}(t) \xrightarrow{t \rightarrow+\infty} 0,-3 \Omega_{0} / 4 \stackrel{t \rightarrow-\infty}{\longleftrightarrow} \Delta(t) \xrightarrow{t \rightarrow+\infty} 3 \Omega_{0} / 4$, $\Omega_{0} / 2 \xrightarrow{t \rightarrow-\infty} \delta(t) \xrightarrow{t \rightarrow+\infty} \Omega_{0} / 2$.

Considering coinciding pulses: $\Omega_{P}(t)=\Omega_{S}(t) \equiv \Omega(t)$ (i.e. $\left.u_{P}(t)=u_{S}(t) \equiv \Omega(t) / 2\right)$, we get expressions for the two-photon detuning and the pulses based on (4.109) and (4.110):

$$
\begin{equation*}
\delta=\frac{\Omega_{0}}{\sqrt{3}} \sqrt{1-\left(\frac{2}{3} \frac{\Delta}{\Omega_{0}}\right)^{2}}, \quad \Omega=\frac{\Omega_{0}}{\sqrt{3}} \sqrt{1-\left(\frac{4}{3} \frac{\Delta}{\Omega_{0}}\right)^{2}} \tag{4.111}
\end{equation*}
$$

considering $\delta(t)$ and $\Omega(t)$ as functions of $\Delta(t)$. Taking for simplicity a monotonic increasing odd function $\Delta(t)=\left(3 \Omega_{0} / 4\right) g(t)$, with $g( \pm \infty)= \pm 1, g(0)=0$, for instance,


Figure 4.10: Time-dependence of dynamics for $\Omega_{0}=14.453 / T$. Upper frame: The control pulses $u_{P}(t)$ and $u_{S}(t)$ (in units of $\left.1 / T\right)$ according to (4.114). Middle frame: Populations with $\psi(t)$ the state solution. Lower frame: The eigenvalues given by (4.108) (in units of $1 / T$ ).
$g(t)=\operatorname{erf}(t / T)$ [12], i.e.

$$
\begin{equation*}
\Delta(t)=\frac{3 \Omega_{0}}{4} \operatorname{erf}\left(\frac{t}{T}\right) \tag{4.112}
\end{equation*}
$$

we obtain

$$
\begin{equation*}
\delta=\frac{\Omega_{0}}{\sqrt{3}} \sqrt{1-\frac{1}{4}\left[\operatorname{erf}\left(\frac{t}{T}\right)\right]^{2}}, \quad \Omega=\frac{\Omega_{0}}{\sqrt{3}} \sqrt{1-\left[\operatorname{erf}\left(\frac{t}{T}\right)\right]^{2}} \tag{4.113}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
u_{P}=u_{S} \equiv \frac{\Omega}{2}=\frac{\Omega_{0}}{2 \sqrt{3}} \sqrt{1-\left[\operatorname{erf}\left(\frac{t}{T}\right)\right]^{2}} \tag{4.114}
\end{equation*}
$$

Numerical implementations are shown in Figure 4.10 for $\Omega_{0}=14.453 / T$, in a situation giving the same energy of the pulses as the one for energy-optimal STIREP $\mathcal{E} \simeq 56 \frac{\hbar}{T}$. We obtain the time area of the transient population in the excited state $A \simeq 0.535 T$, much larger than for energy-optimal STIREP $A \simeq 0.036 T$. Comparing the pulses in Figure 4.9(a), we first notice that the peak in parallel STIRAP $u_{\max } \simeq 4.2 / T$ is roughly equal to one-fifth of the one in energy-optimal STIREP $u_{\max } \simeq 19.6 / T$. Besides, the time duration of the control process is greater than for energy-optimal STIREP.

### 4.2.5 Analogy with a pendulum

In this subsection, we derive the optimal solution in the limit of fully overlapping equal control fields

$$
\begin{equation*}
u_{p}(t)=u_{s}(t)=u(t) \tag{4.115}
\end{equation*}
$$

We show below that the problem becomes analog to the planar pendulum, which is easily solved. We denote by $a(t)$ the area at time $t$ of $u(t)$ :

$$
\begin{equation*}
a(t)=\int_{0}^{t} u\left(t^{\prime}\right) d t^{\prime} \tag{4.116}
\end{equation*}
$$

We have from Eq. (4.5) and Eq. (4.6):

$$
\begin{equation*}
\frac{d}{d t}|x\rangle=u(t)\left(T_{1}+T_{2}\right)|x\rangle \tag{4.117}
\end{equation*}
$$

In this case, the dynamics can be exactly integrated:

$$
\begin{equation*}
|x(t)\rangle=\exp \left[\left(T_{1}+T_{2}\right) a(t)\right]|x(0)\rangle \tag{4.118}
\end{equation*}
$$

A straightforward computation leads to:

$$
\exp \left[\left(T_{1}+T_{2}\right) a(t)\right]=\left[\begin{array}{ccc}
\frac{1+\cos (\sqrt{2} a)}{2} & -\frac{\sin (\sqrt{2} a)}{\sqrt{2}} & \frac{1-\cos (\sqrt{2} a)}{2}  \tag{4.119}\\
\frac{\sin (\sqrt{2} a)}{\sqrt{2}} & \cos (\sqrt{2} a) & -\frac{\sin (\sqrt{2} a)}{\sqrt{2}} \\
\frac{1-\cos (\sqrt{2} a)}{2} & \frac{\sin (\sqrt{2} a)}{\sqrt{2}} & \frac{1+\cos (\sqrt{2} a)}{2}
\end{array}\right]
$$

If the initial point is $|x(0)\rangle=[1,0,0]^{\top}$, we deduce that:

$$
\begin{equation*}
|x(t)\rangle=\left[\frac{1+\cos (\sqrt{2} a)}{2}, \frac{\sin (\sqrt{2} a)}{\sqrt{2}}, \frac{1-\cos (\sqrt{2} a)}{2}\right]^{\top} \tag{4.120}
\end{equation*}
$$

We can verify that the norm of $|x(t)\rangle$ is 1 . The final state is reached if

$$
\begin{equation*}
a(T)=\frac{\pi}{\sqrt{2}} \tag{4.121}
\end{equation*}
$$

where $T$ is the control time.
Minimizing the energy of the controls (4.7) under the constraint of a given loss (4.8) is equivalent of finding the optimal solutions which minimize $2 \int_{0}^{T} u(t)^{2} d t$ while satisfying the constraint condition $2 A=\int_{0}^{T} \sin ^{2}(\sqrt{2} a) d t$. For that purpose, we introduce the Lagrangian

$$
\begin{equation*}
\mathcal{L}_{\lambda}=\dot{a}^{2}+\lambda \sin ^{2}(\sqrt{2} a) \tag{4.122}
\end{equation*}
$$

where $\lambda$ is a Lagrange multiplier. We aim at minimizing $\int_{0}^{T} \mathcal{L}_{\lambda}(t) d t$. A necessary condition is given by the Euler-Lagrange principle:

$$
\begin{equation*}
\ddot{a}-\frac{\lambda}{\sqrt{2}} \sin (2 \sqrt{2} a)=0 \tag{4.123}
\end{equation*}
$$

i.e. the equation of a planar pendulum, with $\lambda$ directly connected to the frequency of the pendulum, $a$ (up to a factor) playing the role of the angle and $u(t)$ of the angular velocity. This equation can be integrated by using Jacobi functions with the conditions $a(0)=0$ and $a(T)=\pi / \sqrt{2}$. The value of $\lambda$ is given by the condition $2 A=\int_{0}^{T} \sin ^{2}(\sqrt{2} a) d t$.

We conclude that, in the case of two equivalent controls, energy-optimal dissipative STIREP can be transformed into a planar pendulum. And the property that the optimal solution corresponds to an oscillating, rotating or separatrix trajectory depends on the value of $A$. According to Subsection 4.2.3, a sufficiently small value of $A$ lead to equivalent controls with the pulse area (4.105): $\int_{0}^{T} u_{p}(t) d t=\int_{0}^{T} u_{s}(t) d t=\pi / \sqrt{2}$, perfectly coinciding with (4.121).
4.3. Optimal control with respect to time constrained by a given admissible loss: time-optimal dissipative STIREP

### 4.3 Optimal control with respect to time constrained by a given admissible loss: time-optimal dissipative STIREP

### 4.3.1 Construction of the pseudo-Hamiltonian

The goal is to derive the optimal solution in the time-minimum case. The parameter $A$ in (4.8) is set to a given value as before and the time $T$ is unfixed. The optimal control problem consists in reaching exactly the target state $\left(x_{1}\left(t_{f}\right)=x_{2}\left(t_{f}\right)=0, x_{3}\left(t_{f}\right)=1\right)$ while minimizing the functional

$$
\begin{equation*}
J=\int_{0}^{T} d t \tag{4.124}
\end{equation*}
$$

under the constraint (3.98):

$$
\begin{equation*}
u_{1}^{2}(t)+u_{2}^{2}(t) \leq u_{0}^{2}, \tag{4.125}
\end{equation*}
$$

where $u_{0}$ is the maximum field intensity according to (3.109). The pesudo Hamiltonian reads (with the standard choice $p_{0}=1 / 2$ )

$$
\begin{align*}
\tilde{H}_{c} & =\left[\lambda_{1}, \lambda_{2}, \lambda_{3}\right]\left[\begin{array}{c}
-u_{p} x_{2} \\
u_{p} x_{1}-u_{s} x_{3} \\
u_{s} x_{2}
\end{array}\right]+\mu x_{2}^{2}-\frac{1}{2} \\
& =u_{p}\left(\lambda_{2} x_{1}-\lambda_{1} x_{2}\right)+u_{s}\left(\lambda_{3} x_{2}-\lambda_{2} x_{3}\right)+\mu x_{2}^{2}-\frac{1}{2} . \tag{4.126}
\end{align*}
$$

Subtracting the constant $p_{0}$, we rewrite the pesudo Hamiltonian:

$$
\begin{equation*}
H_{c} \equiv \tilde{H}_{c}+\frac{1}{2}=u_{p}\left(\lambda_{2} x_{1}-\lambda_{1} x_{2}\right)+u_{s}\left(\lambda_{3} x_{2}-\lambda_{2} x_{3}\right)+\mu x_{2}^{2}, \tag{4.127}
\end{equation*}
$$

where the costate $\Lambda$ has four components $\Lambda=\left[\lambda^{\top}, \mu\right]^{\top}$ with $\lambda=\left[\lambda_{1}, \lambda_{2}, \lambda_{3}\right]^{\top}$. The adjoint equations of the costate read

$$
\begin{align*}
\dot{\lambda}_{1} & =-\frac{\partial H_{c}}{\partial x_{1}}=-\lambda_{2} u_{p},  \tag{4.128a}\\
\dot{\lambda}_{2} & =-\frac{\partial H_{c}}{\partial x_{2}}=\lambda_{1} u_{p}-\lambda_{3} u_{s}-2 \mu x_{2},  \tag{4.182b}\\
\dot{\lambda}_{3} & =-\frac{\partial H_{c}}{\partial x_{3}}=\lambda_{2} u_{s},  \tag{4.128c}\\
\dot{\mu} & =0 \quad \text { i.e. } \quad \mu=\text { const. } \tag{4.128d}
\end{align*}
$$

The pseudo Hamiltonian is of the form (3.95) $H_{c}=H_{0}+u_{p} H_{p}+u_{s} H_{s}$ (see Section 3.5), with the control variable $u_{p}$ and $u_{s}$ and $H_{p}=\lambda_{2} x_{1}-\lambda_{1} x_{2}, H_{s}=\lambda_{3} x_{2}-\lambda_{2} x_{3}$, we can thus apply the results of this Section 3.5:

$$
\begin{align*}
& u_{p}=\left(\lambda_{2} x_{1}-\lambda_{1} x_{2}\right) / R,  \tag{4.129a}\\
& u_{s}=\left(\lambda_{3} x_{2}-\lambda_{2} x_{3}\right) / R, \tag{4.129b}
\end{align*}
$$

with

$$
\begin{equation*}
R=\sqrt{\left(\lambda_{2} x_{1}-\lambda_{1} x_{2}\right)^{2}+\left(\lambda_{3} x_{2}-\lambda_{2} x_{3}\right)^{2}} \tag{4.130}
\end{equation*}
$$

This leads to

$$
\begin{equation*}
H_{c}=R+\mu x_{2}^{2} \tag{4.131}
\end{equation*}
$$

and the controls attain the maximum of the constraint at each time

$$
\begin{equation*}
u_{p}^{2}+u_{s}^{2}=u_{0}^{2} . \tag{4.132}
\end{equation*}
$$

We can make a change of variables for the time renormalizing the field amplitude as

$$
\begin{align*}
\tilde{u}_{p} & =\frac{u_{p}}{\sqrt{u_{p}^{2}+u_{s}^{2}}}=\frac{u_{p}}{u_{0}}  \tag{4.133a}\\
\tilde{u}_{s} & =\frac{u_{s}}{\sqrt{u_{p}^{2}+u_{s}^{2}}}=\frac{u_{s}}{u_{0}}  \tag{4.133b}\\
\tilde{t} & =u_{0} t \tag{4.133c}
\end{align*}
$$

i.e.

$$
\begin{equation*}
\tilde{u}_{p}^{2}+\tilde{u}_{s}^{2}=1 \tag{4.134}
\end{equation*}
$$

and the equation becomes:

$$
\frac{d}{d \tilde{t}}|\tilde{x}\rangle=\tilde{A}|\tilde{x}\rangle, \quad|\tilde{x}(\tilde{t})\rangle \equiv|x(t)\rangle, \quad \tilde{A}=\left[\begin{array}{ccc}
0 & -\tilde{u}_{p} & 0  \tag{4.135}\\
\tilde{u}_{p} & 0 & -\tilde{u}_{s} \\
0 & \tilde{u}_{s} & 0
\end{array}\right]
$$

This means that we can always renormalize the field amplitudes by modifying the optimal time accordingly. We will consider below the tilde variables (corresponding to finding the optimal time for $u_{0}=1$ ).

Introducing angle coordinates:

$$
\begin{align*}
& x_{1}=\cos \varphi \cos \theta  \tag{4.136a}\\
& x_{2}=\sin \varphi  \tag{4.136b}\\
& x_{3}=\cos \varphi \sin \theta \tag{4.136c}
\end{align*}
$$

with the initial condition $\varphi\left(t_{i}\right)=0, \theta\left(t_{i}\right)=0$ and the final condition $\varphi\left(t_{f}\right)=0$, $\theta\left(t_{f}\right)=\pi / 2$. The equations of the dynamics (4.135)

$$
\begin{align*}
\dot{\varphi} & =\tilde{u}_{p} \cos \theta-\tilde{u}_{s} \sin \theta  \tag{4.137a}\\
\dot{\theta} & =\left(\tilde{u}_{p} \sin \theta+\tilde{u}_{s} \cos \theta\right) \tan \varphi \tag{4.137b}
\end{align*}
$$

can be simplified as

$$
\begin{align*}
\dot{\varphi} & =\tilde{v}_{p}  \tag{4.138a}\\
\dot{\theta} & =-\tilde{v}_{s} \tan \varphi, \tag{4.138b}
\end{align*}
$$

after a rotation on the control fields

$$
\left[\begin{array}{c}
\tilde{v}_{p}  \tag{4.139}\\
\tilde{v}_{s}
\end{array}\right]=\left[\begin{array}{cc}
\cos \theta & -\sin \theta \\
-\sin \theta & -\cos \theta
\end{array}\right]\left[\begin{array}{c}
\tilde{u}_{p} \\
\tilde{u}_{s}
\end{array}\right]
$$

leading to an invariant cost on the new field variables since $\tilde{u}_{p}^{2}+\tilde{u}_{s}^{2}=\tilde{v}_{p}^{2}+\tilde{v}_{s}^{2}=1$. We arrive at:

$$
\begin{equation*}
H_{c}=\lambda_{\varphi} \dot{\varphi}+\lambda_{\theta} \dot{\theta}+\mu \sin ^{2} \varphi=\lambda_{\varphi} \tilde{v}_{p}-\lambda_{\theta} \tilde{v}_{s} \tan \varphi+\mu \sin ^{2} \varphi \tag{4.140}
\end{equation*}
$$

with $\Lambda=\left[\lambda_{\varphi}, \lambda_{\theta}, \mu\right]^{\top}$ the costate gathering the conjugate momenta of $\varphi, \theta$, and $y$, respectively. The Hamilton equations

$$
\begin{align*}
\dot{\varphi} & =\frac{\partial H_{c}}{\partial \lambda_{\varphi}}  \tag{4.141a}\\
\dot{\theta} & =\frac{\partial H_{c}}{\partial \lambda_{\theta}} \tag{4.141b}
\end{align*}
$$

lead to the equation of motion (4.138) and to

$$
\begin{align*}
\dot{\lambda}_{\varphi} & =-\frac{\partial H_{c}}{\partial \varphi}=\frac{\tilde{v}_{s}}{\cos ^{2} \varphi} \lambda_{\theta}-\mu \sin (2 \varphi)  \tag{4.142a}\\
\dot{\lambda}_{\theta} & =-\frac{\partial H_{c}}{\partial \theta}=0  \tag{4.142b}\\
\dot{\mu} & =-\frac{\partial H_{c}}{\partial y}=0 \tag{4.142c}
\end{align*}
$$

This implies that $\lambda_{\theta}$ and $\mu$ are constants of motion. $H_{c}$ (4.140) is again of the form (3.95) implying:

$$
\begin{equation*}
H_{c}=\sqrt{\lambda_{\varphi}^{2}+\lambda_{\theta}^{2} \tan ^{2} \varphi}+\mu \sin ^{2} \varphi \tag{4.143}
\end{equation*}
$$

and

$$
\begin{equation*}
\tilde{v}_{p}=\frac{\lambda_{\varphi}}{R}, \quad \tilde{v}_{s}=-\frac{\lambda_{\theta} \tan \varphi}{R} \tag{4.144}
\end{equation*}
$$

with

$$
\begin{equation*}
R=\sqrt{\lambda_{\varphi}^{2}+\lambda_{\theta}^{2} \tan ^{2} \varphi} \tag{4.145}
\end{equation*}
$$

And the equations of motion read:

$$
\begin{align*}
\dot{\varphi} & =\frac{\lambda_{\varphi}}{R}  \tag{4.146a}\\
\dot{\theta} & =\frac{\lambda_{\theta} \tan ^{2} \varphi}{R}  \tag{4.146b}\\
\dot{y} & =\sin ^{2} \varphi  \tag{4.146c}\\
\dot{\lambda}_{\varphi} & =-\lambda_{\theta}^{2} \frac{\sin \varphi}{R \cos ^{3} \varphi}-\mu \sin (2 \varphi) \tag{4.146~d}
\end{align*}
$$

with the boundary conditions

$$
\begin{align*}
& \varphi\left(t_{i}\right)=0, \quad \theta\left(t_{i}\right)=0, \quad y\left(t_{i}\right)=0  \tag{4.147a}\\
& \varphi\left(t_{f}\right)=0, \quad \theta\left(t_{f}\right)=\pi / 2, \quad y\left(t_{f}\right)=A \tag{4.147b}
\end{align*}
$$

### 4.3.2 Construction of the optimal trajectories from the Pontryagin maximum principle

Since $H_{c}=h$ is a constant, we get

$$
\begin{equation*}
\lambda_{\varphi}= \pm \sqrt{\left(h-\mu \sin ^{2} \varphi\right)^{2}-\lambda_{\theta}^{2} \tan ^{2} \varphi} \tag{4.148}
\end{equation*}
$$

Following the same lines as in the energy minimum case, we have:

$$
\begin{array}{r}
\dot{\varphi}=\frac{\lambda_{\varphi}}{R}= \pm \frac{\sqrt{\left(h-\mu \sin ^{2} \varphi\right)^{2}-\lambda_{\theta}^{2} \tan ^{2} \varphi}}{h-\mu \sin ^{2} \varphi}, \\
\frac{d \varphi}{d \theta}=\frac{\dot{\varphi}}{\dot{\theta}}= \pm \frac{\sqrt{\left(h-\mu \sin ^{2} \varphi\right)^{2}-\lambda_{\theta}^{2} \tan ^{2} \varphi}}{\lambda_{\theta} \tan ^{2} \varphi} . \tag{4.149b}
\end{array}
$$

One can normalize the constants $\mu$ and $\lambda_{\theta}$ as

$$
\begin{equation*}
\tilde{\mu}=\frac{\mu}{h}, \quad \tilde{\lambda}_{\theta}=\frac{\lambda_{\theta}}{h}, \tag{4.150}
\end{equation*}
$$

therefore, Eq.(4.149) is transformed into

$$
\begin{align*}
\dot{\varphi} & = \pm \frac{\sqrt{\left(1-\tilde{\mu} \sin ^{2} \varphi\right)^{2}-\tilde{\lambda}_{\theta}^{2} \tan ^{2} \varphi}}{1-\tilde{\mu} \sin ^{2} \varphi}  \tag{4.151a}\\
\frac{d \varphi}{d \theta} & = \pm \frac{\sqrt{\left(1-\tilde{\mu} \sin ^{2} \varphi\right)^{2}-\tilde{\lambda}_{\theta}^{2} \tan ^{2} \varphi}}{\tilde{\lambda}_{\theta} \tan ^{2} \varphi} \tag{4.151b}
\end{align*}
$$

We know that $\varphi$ has to reach its maximum value at $t=T / 2$, with $T$ the control time to be determined optimally:

$$
\begin{equation*}
\dot{\varphi}(T / 2)=0 \tag{4.152}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
\left(1-\tilde{\mu} \sin ^{2} \varphi_{0}\right)^{2}=\tilde{\lambda}_{\theta}^{2} \tan ^{2} \varphi_{0} \tag{4.153}
\end{equation*}
$$

with the notation $\varphi_{0}=\varphi(T / 2)$. This equation shows a dependence upon $\lambda_{\theta}^{2}$. We can thus limit our study for positive $\lambda_{\theta}$.

When $\tilde{\mu} \neq 0$, and multiplying Eq. (4.153) by $1-\sin ^{2} \varphi_{0}$, we obtain that $\sin ^{2} \varphi_{0}$ is a (positive and less than (or equal to) one) root of a cubic polynomias:

$$
\begin{equation*}
1-\left(1+\tilde{\lambda}_{\theta}^{2}+2 \tilde{\mu}\right) \sin ^{2} \varphi_{0}+\left(\tilde{\mu}^{2}+2 \tilde{\mu}\right) \sin ^{4} \varphi_{0}-\tilde{\mu}^{2} \sin ^{6} \varphi_{0}=0, \tag{4.154}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
1-\left(1+\tilde{\lambda}_{\theta}^{2}+2 \tilde{\mu}\right) X+\left(\tilde{\mu}^{2}+2 \tilde{\mu}\right) X^{2}-\tilde{\mu}^{2} X^{3}=0 \tag{4.155}
\end{equation*}
$$

with $X=\sin ^{2} \varphi_{0}$. It is important to note that multiplying Eq. (4.153) by $1-\sin ^{2} \varphi_{0}$ introduces a root $\varphi_{0}=\pi / 2$, except for $\tilde{\lambda}_{\theta}=0$ and $\tilde{\mu}=1$ for which $\varphi_{0}=\pi / 2$ is a true root of (4.153). We analyze the solution $\varphi_{0}$ near $\pi / 2$ by setting $\varphi_{0}=\pi / 2+\varepsilon,|\varepsilon| \ll 1$,
into (4.153), which gives a relation between $\tilde{\lambda}_{\theta}$ and $\tilde{\mu}$ for a given $\varepsilon$ :

$$
\begin{equation*}
\left|\tilde{\lambda}_{\theta}\right|=|\varepsilon| \times|1-\tilde{\mu}|, \tag{4.156}
\end{equation*}
$$

which shows how $\tilde{\lambda}_{\theta}$ goes to zero and $\varphi_{0}$ to $\pi / 2$ for a given $\tilde{\mu}$.
When $\tilde{\mu}=0$, we obtain

$$
\begin{equation*}
\varphi_{0}=\arctan \frac{1}{\tilde{\lambda}_{\theta}} \tag{4.157}
\end{equation*}
$$

One can alternatively use analytic Cardano's method to solve equation (4.155), as shown below. Substituting

$$
\begin{equation*}
X=Y+\frac{\tilde{\mu}^{2}+2 \tilde{\mu}}{3 \tilde{\mu}^{2}}=Y+\frac{\tilde{\mu}+2}{3 \tilde{\mu}} \tag{4.158}
\end{equation*}
$$

Eq. (4.155) can be depressed to

$$
\begin{equation*}
Y^{3}+p Y+q=0 \tag{4.159}
\end{equation*}
$$

with $p=\frac{3 \tilde{\lambda}_{\theta}^{2}-1+2 \tilde{\mu}-\tilde{\mu}^{2}}{3 \tilde{\mu}^{2}}, q=\frac{2+9(\tilde{\mu}+2) \tilde{\lambda}_{\theta}^{2}-6 \tilde{\mu}+6 \tilde{\mu}^{2}-2 \tilde{\mu}^{3}}{27 \tilde{\mu}^{3}}$. We define the discriminant of Eq. (4.159) as

$$
\begin{equation*}
\Delta=\left(\frac{q}{2}\right)^{2}+\left(\frac{p}{3}\right)^{3} \tag{4.160}
\end{equation*}
$$

When $\Delta>0$, the solutions of Eq. (4.159) read (with only the first one being real)

$$
\begin{align*}
& Y_{1}=\sqrt[3]{-\frac{q}{2}+\sqrt{\left(\frac{q}{2}\right)^{2}+\left(\frac{p}{3}\right)^{3}}}+\sqrt[3]{-\frac{q}{2}-\sqrt{\left(\frac{q}{2}\right)^{2}+\left(\frac{p}{3}\right)^{3}}}  \tag{4.161a}\\
& Y_{2}=\omega \sqrt[3]{-\frac{q}{2}+\sqrt{\left(\frac{q}{2}\right)^{2}+\left(\frac{p}{3}\right)^{3}}}+\omega^{2} \sqrt[3]{-\frac{q}{2}-\sqrt{\left(\frac{q}{2}\right)^{2}+\left(\frac{p}{3}\right)^{3}}}  \tag{4.161b}\\
& Y_{3}=\omega^{2} \sqrt[3]{-\frac{q}{2}+\sqrt{\left(\frac{q}{2}\right)^{2}+\left(\frac{p}{3}\right)^{3}}}+\omega \sqrt[3]{-\frac{q}{2}-\sqrt{\left(\frac{q}{2}\right)^{2}+\left(\frac{p}{3}\right)^{3}}} \tag{4.161c}
\end{align*}
$$

with

$$
\begin{equation*}
\omega=e^{2 i \pi / 3} \tag{4.162}
\end{equation*}
$$

and where the cube root is used (imposing a real argument).
When $\Delta \leq 0$, which implies that $p$ is negative, the solutions of Eq. (4.159) read (being all real):

$$
\begin{equation*}
Y_{k+1}=2 \sqrt{\frac{-p}{3}} \cos \left(\frac{1}{3} \arccos \left(\frac{3 q}{2 p} \sqrt{\frac{3}{-p}}\right)+\frac{2 k \pi}{3}\right) \quad \text { with } \quad k \in\{0,1,2\} \tag{4.163}
\end{equation*}
$$

The relationship between the roots and the coefficients is

$$
\begin{align*}
Y_{1}+Y_{2}+Y_{3} & =0  \tag{4.164a}\\
\frac{1}{Y_{1}}+\frac{1}{Y_{2}}+\frac{1}{Y_{3}} & =-\frac{p}{q}  \tag{4.164b}\\
Y_{1} Y_{2} Y_{3} & =-q \tag{4.164c}
\end{align*}
$$

When $\Delta<0$, Eq. (4.159) has three different real roots. When $\Delta=0$ (implying $p \leq 0$ ), Eq. (4.159) has two roots when $p, q \neq 0$ :

$$
\begin{align*}
& Y_{1}=2 \sqrt[3]{-\frac{q}{2}}  \tag{4.165a}\\
& Y_{2}=\sqrt[3]{\frac{q}{2}}=Y_{3} \tag{4.165b}
\end{align*}
$$

and three being equal to zero when $q=0$ (implying $p=0$ ).

We are searching for positive and less than (or equal to) one real roots. We first determine the sign of the discriminant:

$$
\begin{align*}
\Delta & =\frac{1}{27^{2} \tilde{\mu}^{6}}\left[\left(\frac{9}{2}(\tilde{\mu}+2) \tilde{\lambda}_{\theta}^{2}+1-3 \tilde{\mu}+3 \tilde{\mu}^{2}-\tilde{\mu}^{3}\right)^{2}+\left(3 \tilde{\lambda}_{\theta}^{2}-1+2 \tilde{\mu}-\tilde{\mu}^{2}\right)^{3}\right] \\
& =\frac{\tilde{\lambda}_{\theta}^{2}}{27 \tilde{\mu}^{6}}\left[\tilde{\lambda}_{\theta}^{4}+\left(-\frac{1}{4} \tilde{\mu}^{2}+5 \tilde{\mu}+2\right) \tilde{\lambda}_{\theta}^{2}+1-3 \tilde{\mu}+3 \tilde{\mu}^{2}-\tilde{\mu}^{3}\right] \\
& =\frac{\tilde{\lambda}_{\theta}^{2}}{27 \tilde{\mu}^{6}} P_{2}\left(\tilde{\lambda}_{\theta}^{2}\right) . \tag{4.166}
\end{align*}
$$

The sign of $\Delta$ is given by the sign of the second-order polynomial $P_{2}\left(\tilde{\lambda}_{\theta}^{2}\right)$ in $\tilde{\lambda}_{\theta}^{2}$ (note that the roots of this polynomials have thus to be positive). We first note that $\Delta=0$ when $\tilde{\lambda}_{\theta}=0$.

The discriminant associated to this polynomial $P_{2}\left(\tilde{\lambda}_{\theta}^{2}\right)$ writes

$$
\begin{align*}
\Delta^{\prime} & =\left(-\frac{1}{4} \tilde{\mu}^{2}+5 \tilde{\mu}+2\right)^{2}-4\left(1-3 \tilde{\mu}+3 \tilde{\mu}^{2}-\tilde{\mu}^{3}\right) \\
& =\frac{1}{16} \tilde{\mu}(\tilde{\mu}+8)^{3} . \tag{4.167}
\end{align*}
$$

When $\tilde{\mu} \in[-8,0]$, i.e. $\Delta^{\prime} \leq 0$, we have $P_{2}\left(\tilde{\lambda}_{\theta}^{2}\right) \geq 0$, i.e. $\Delta \geq 0$, for all $\tilde{\lambda}_{\theta}$.
We have $P_{2}\left(\tilde{\lambda}_{\theta}^{2}\right)=0$, i.e. $\Delta=0$, for $\tilde{\mu}=-8$ and $\tilde{\lambda}_{\theta}= \pm \sqrt{27}$ (the other possibility $\tilde{\mu}=0$ leads to the incompatible solution $\tilde{\lambda}_{\theta}^{2}=-1$ ).

When $\tilde{\mu} \in]-\infty,-8[\cup] 0,+\infty\left[\right.$, i.e. $\Delta^{\prime}>0$, there are two roots for $P_{2}\left(\tilde{\lambda}_{\theta}^{2}\right)$ :

$$
\begin{equation*}
\tilde{\lambda}_{\theta, \pm}^{2}=\frac{1}{2}\left(\frac{1}{4} \tilde{\mu}^{2}-5 \tilde{\mu}-2 \pm \frac{1}{4} \sqrt{\tilde{\mu}(\tilde{\mu}+8)^{3}}\right) \tag{4.168}
\end{equation*}
$$

When $\tilde{\mu}<-8$, these roots are both positive, and thus compatible. In this case, $\Delta<0$ for $\tilde{\lambda}_{\theta,-}^{2}<\tilde{\lambda}_{\theta}^{2}<\tilde{\lambda}_{\theta,+}^{2}$, i.e. for $\tilde{\lambda}_{\theta,-}<\tilde{\lambda}_{\theta}<\tilde{\lambda}_{\theta,+}$ or for $-\tilde{\lambda}_{\theta,+}<\tilde{\lambda}_{\theta}<-\tilde{\lambda}_{\theta,-} ; \Delta \geq 0$ for the other ranges of $\left.\left.\tilde{\lambda}_{\theta}: \tilde{\lambda}_{\theta} \in\right]-\infty,-\tilde{\lambda}_{\theta,+}\right] \cup\left[-\tilde{\lambda}_{\theta,-}, \tilde{\lambda}_{\theta,-}\right] \cup\left[\tilde{\lambda}_{\theta,+},+\infty[\right.$.
When $\tilde{\mu}>0$, only $\tilde{\lambda}_{\theta,+}^{2} \geq 0$ is compatible and when $\tilde{\mu} \geq 1$. We have thus to distinguish two ranges for $\tilde{\mu}$ :
For $\tilde{\mu} \in] 0,1]$, we have $P_{2}\left(\tilde{\lambda}_{\theta}^{2}\right) \geq 0$ since the two roots are negative, i.e. $\Delta \geq 0$, for all $\tilde{\lambda}_{\theta}$. We have $\Delta=0$ for $\tilde{\mu}=1$ and $\tilde{\lambda}_{\theta}=0$. In fact, $\Delta=0$ for all $\tilde{\lambda}_{\theta}=0$ according to the definition of $\Delta$ (4.166).
For $\tilde{\mu}>1$, we have $P_{2}\left(\tilde{\lambda}_{\theta}^{2}\right)>0$, i.e. $\Delta>0$, for $\tilde{\lambda}_{\theta}^{2}>\tilde{\lambda}_{\theta,+}^{2}$, i.e. for $\tilde{\lambda}_{\theta}>\tilde{\lambda}_{\theta,+}$ or $\tilde{\lambda}_{\theta}<-\tilde{\lambda}_{\theta,+}$. We have $\Delta \leq 0$ for $-\tilde{\lambda}_{\theta,+} \leq \tilde{\lambda}_{\theta} \leq \tilde{\lambda}_{\theta,+}$.

When $\Delta>0$, the real solution reads

$$
\begin{equation*}
\sin ^{2} \varphi_{0}=\sqrt[3]{-\frac{q}{2}+\frac{\tilde{\lambda}_{\theta}}{\sqrt{27} \tilde{\mu}^{3}} \sqrt{P_{2}\left(\tilde{\lambda}_{\theta}^{2}\right)}}+\sqrt[3]{-\frac{q}{2}-\frac{\tilde{\lambda}_{\theta}}{\sqrt{27} \tilde{\mu}^{3}} \sqrt{P_{2}\left(\tilde{\lambda}_{\theta}^{2}\right)}}+\frac{\tilde{\mu}+2}{3 \tilde{\mu}} \tag{4.169}
\end{equation*}
$$

When $\Delta=0$, the two solutions read

$$
\begin{align*}
\sin ^{2} \varphi_{0} & =2 \sqrt[3]{-\frac{q}{2}}+\frac{\tilde{\mu}+2}{3 \tilde{\mu}}  \tag{4.170a}\\
\sin ^{2} \varphi_{0} & =-\sqrt[3]{-\frac{q}{2}}+\frac{\tilde{\mu}+2}{3 \tilde{\mu}}=\sqrt[3]{\frac{q}{2}}+\frac{\tilde{\mu}+2}{3 \tilde{\mu}} \tag{4.170b}
\end{align*}
$$

When $\Delta<0$, the three real solutions read (with $k \in\{0,1,2\}$ )

$$
\begin{equation*}
\sin ^{2} \varphi_{0}=2 \sqrt{\frac{-p}{3}} \cos \left(\frac{1}{3} \arccos \left(\frac{3 q}{2 p} \sqrt{\frac{3}{-p}}\right)+\frac{2 k \pi}{3}\right)+\frac{\tilde{\mu}+2}{3 \tilde{\mu}} \tag{4.171}
\end{equation*}
$$

In summary, we conclude that we have the following real roots:

- when $\tilde{\lambda}_{\theta}=0$, according to (4.153),
- one root, when $\tilde{\mu} \geq 1$ :

$$
\begin{equation*}
\sin ^{2} \varphi_{0}=\frac{1}{\tilde{\mu}} \tag{4.172}
\end{equation*}
$$

- no root, when $\tilde{\mu}<1$;
- when $\tilde{\lambda}_{\theta} \neq 0$ :
- for $\tilde{\mu} \leq-8$,
one root (4.169), for $\left.\tilde{\lambda}_{\theta} \in\right] 0, \tilde{\lambda}_{\theta,-}[U] \tilde{\lambda}_{\theta,+},+\infty[$,
two roots (4.170), for $\tilde{\lambda}_{\theta}=\tilde{\lambda}_{\theta,-}$ or $\tilde{\lambda}_{\theta}=\tilde{\lambda}_{\theta,+}$,
three roots (4.171), for $\tilde{\lambda}_{\theta,-}<\tilde{\lambda}_{\theta}<\tilde{\lambda}_{\theta,+}$;
- for $\tilde{\mu} \in]-8,1]$, one root (4.169) (which coincides with (4.157) for $\tilde{\mu}=0$ );
- for $\tilde{\mu}>1$,
one root (4.169), for $\tilde{\lambda}_{\theta}>\tilde{\lambda}_{\theta,+}$,
two roots (4.170), for $\tilde{\lambda}_{\theta}=\tilde{\lambda}_{\theta,+}$ (which become one root: $\varphi_{0}=\pi / 2$ when $\tilde{\mu}=1\left(\right.$ giving then $\left.\left.\tilde{\lambda}_{\theta}=0\right)\right)$,
three roots (4.171), for $0<\tilde{\lambda}_{\theta}<\tilde{\lambda}_{\theta,+}$.

One can analyze the limit when $\tilde{\lambda}_{\theta}$ goes to zero:

In the range $\tilde{\mu} \leq 1$, the root given by (4.169), where, for $\tilde{\lambda}_{\theta}=0$

$$
\begin{equation*}
q=\frac{2}{27 \tilde{\mu}^{3}}(1-\tilde{\mu})^{3}, \quad p=-\frac{1}{3} \frac{(1-\tilde{\mu})^{2}}{\tilde{\mu}^{2}} \tag{4.173}
\end{equation*}
$$

gives

$$
\begin{equation*}
\sin ^{2} \varphi_{0}=1, \tag{4.174}
\end{equation*}
$$

which is compatible with $\varphi_{0}$ going to $\pi / 2$ for $\tilde{\lambda}_{\theta}$ going to 0 .

In the range $\tilde{\mu}>1$, Eq. (4.171):

$$
\begin{align*}
\sin ^{2} \varphi_{0} & =\frac{2}{3} \frac{|1-\tilde{\mu}|}{|\tilde{\mu}|} \cos \left(\frac{1}{3} \arccos \left(\operatorname{sgn}\left(\frac{\tilde{\mu}-1}{\tilde{\mu}}\right)\right)+\frac{2 k \pi}{3}\right)+\frac{\tilde{\mu}+2}{3 \tilde{\mu}}  \tag{4.175}\\
& =\frac{2}{3} \frac{\tilde{\mu}-1}{\tilde{\mu}} \cos \left(\frac{2 k \pi}{3}\right)+\frac{\tilde{\mu}+2}{3 \tilde{\mu}}
\end{align*}
$$

gives the two roots

$$
\begin{equation*}
\sin ^{2} \varphi_{0}=\{1,1 / \tilde{\mu}\} \tag{4.176}
\end{equation*}
$$



Figure 4.11: Contour plots of solution(s) of $\varphi_{0}$ from Eq.(4.153) as a function of $\tilde{\mu}$ and $\tilde{\lambda}_{\theta}$. Absence of solutions is indicated in black.
4.3. Optimal control with respect to time constrained by a given admissible loss: time-optimal dissipative STIREP

Contour plots as a function of $\tilde{\mu}$ and $\tilde{\lambda}_{\theta}$ shown in Figure 4.11 demonstrate the solution(s) of $\varphi_{0}$ satisfying (4.153). Considering that the maximum number of solutions is three, we make three plots. Absence of solutions is indicated by black color ("0"). In addition, in the situation of one solution, we save it in Figure 4.11(a) and record the two others as zero in Figure 4.11(b) and 4.11(c); in the situation of two solutions, we save them in Figure 4.11(a) and 4.11(b) and set the last one as zero in Figure 4.11(c).

(a)

(b)

Figure 4.12: Contour plots of logarithm to the base 10 of the absolute values of the difference between $\varphi(\pi / 4)$ (from Eq. (4.151b)) and $\varphi_{0}$ (from Eq. (4.153)), as a function of $\tilde{\mu}$ and $\tilde{\lambda}_{\theta}$ for (a) $\tilde{\mu} \leq-8$; and (b) $\left.\left.\tilde{\mu} \in\right]-8,1\right]$.

Solving the differential equation (4.151b) numerically (with Matlab) and determining $\varphi(\theta=\pi / 4)$, we make contour plots of $\left|\varphi(\pi / 4)-\varphi_{0}\right|$ as a function of $\tilde{\mu}$ and $\tilde{\lambda}_{\theta}$ in logarithmic scale, as shown in Figure 4.12. The goal is to find a first approximation of the values of $\tilde{\mu}$ and $\tilde{\lambda}_{\theta}$ satisfying $\left|\varphi(\pi / 4)-\varphi_{0}\right|=0$, which corresponds to $\theta\left(\varphi_{0}\right)=\pi / 4$. We note that for $\tilde{\mu} \leq-8,\left|\varphi(\pi / 4)-\varphi_{0}\right|=0$ only appears for the second root of $\varphi_{0}$ (i.e. (4.171) when $k=1$ ) in the case of three roots for Eq. (4.153) (see Figure 4.12(a)) and the solution exists very close to the critical value of $\Delta<0$. In addition, we emphasize that the results become strongly sensitive to $\tilde{\lambda}_{\theta}$ for large negative $\tilde{\mu}$, which explains why we keep $\tilde{\lambda}_{\theta}$ with different accuracy. For $\left.\left.\tilde{\mu} \in\right]-8,1\right]$, there is one root of $\varphi_{0}$ (i.e. (4.169)), and $\left|\varphi(\pi / 4)-\varphi_{0}\right|=0$ always holds (see Figure $4.12(\mathrm{~b})$ ). For $\tilde{\mu}>1$, no solution exists for $\left|\varphi(\pi / 4)-\varphi_{0}\right|=0$. The black curves in Figure 4.12 show rough acceptable solutions (whose accuracy is here limited by the discretization of $\tilde{\mu}$ and $\tilde{\lambda}_{\theta}$ ). We improve the accuracy of the values of $\tilde{\lambda}_{\theta}$ for a given $\tilde{\mu}$ around the black curve found previously, and the obtained data are shown in Table 4.3. We could not obtain the results for larger negative $\tilde{\mu}$, because the sensitivity of the results requires more precise values of $\tilde{\lambda}_{\theta}$ which are beyond the limits of the effective numerical settings of Matlab. We observe that $\tilde{\lambda}_{\theta}$ decreases monotonically for increasing $\tilde{\mu}$, Figure 4.13 showing the dependence of these two parameters.

| $\tilde{\mu}$ | $\tilde{\lambda}_{\theta}$ | $\log _{10}\left\|\varphi(\pi / 4)-\varphi_{0}\right\|$ |
| :---: | :---: | :---: |
| 0.5 | 0.25879 | -12.455177644518727 |
| 0 | 0.57735 | -13.827484971826197 |
| -0.5 | 0.90476 | -11.946586098350844 |
| -1 | 1.23456 | -13.134388310705363 |
| -1.5 | 1.56434 | -11.870156493404458 |
| -2 | 1.89263 | -12.460852967914164 |
| -2.5 | 2.21825 | -12.004955846391741 |
| -3 | 2.53997 | -13.567199943852273 |
| -3.5 | 2.85642 | -12.635483710881227 |
| -4 | 3.16595 | -11.913394536494193 |
| -4.5 | 3.46665 | -11.218177330794603 |
| -5 | 3.75639 | -12.667684417218629 |
| -5.5 | 4.03322 | -11.614661562412490 |
| -6 | 4.29565 | -12.985640089209660 |
| -6.5 | 4.543113 | -12.322740308031186 |
| -7 | 4.776073 | -12.010848004359689 |
| -7.5 | 4.995772 | -12.910050009798592 |


| -8 | 5.203814 | -12.525726361891747 |
| :---: | :---: | :---: |
| -8.5 | 5.401802 | -10.053200113074293 |
| -9 | 5.591159 | -11.046630512135870 |
| -9.5 | 5.773058 | -11.083922558606149 |
| -10 | 5.948450 | -10.793101979103550 |
| -10.5 | 6.1180957 | -10.648733450827898 |
| -11 | 6.2826117 | -11.030475961167475 |
| -11.5 | 6.4425007 | -10.829212617518843 |
| -12 | 6.5981805 | -10.842487715420113 |
| -12.5 | 6.75000296 | -11.202026623410891 |
| -13 | 6.89826843 | -10.939429181948508 |
| -13.5 | 7.04323635 | -10.908617791130567 |
| -14 | 7.185133286 | -10.362734131753392 |
| -14.5 | 7.324158901 | -10.603082380660583 |
| -15 | 7.460490528 | -11.202222444405354 |
| -15.5 | 7.5942867762 | -10.674278932950855 |
| -16 | 7.7256903690 | -10.237438317870252 |
| -16.5 | 7.8548304148 | -11.586758808237050 |
| -17 | 7.98182426212 | -10.453982848125017 |
| -17.5 | 8.10677902312 | -11.468293922237317 |
| -18 | 8.22979283925 | -11.406759705012481 |
| -18.5 | 8.35095594416 | -11.058805462796315 |
| -19 | 8.47035156274 | -11.916924276840200 |
| -19.5 | 8.588056677662 | -10.635722273841216 |
| -20 | 8.704142687429 | -10.259381583932900 |
| -20.5 | 8.8186759747537 | -11.410922081662752 |
| -21 | 8.9317184002888 | -10.366964381960122 |
| -21.5 | 9.0433277337173 | -10.031027661858733 |
| -22 | 9.1535580319427 | -10.069415484038799 |
| -22.5 | 9.262459972324599 | -11.162346957138606 |
| -23 | 9.370081147493655 | -10.032442458984171 |
| -23.5 | 9.476466327158352 | -9.432913468809332 |
| -24 | 9.581657691415900 | -9.770536553920516 |
| -24.5 | 9.685695039350745 | -9.890509238277009 |

Table 4.3: Values of couple $\left(\tilde{\mu}, \tilde{\lambda}_{\theta}\right)$ satisfying $\left|\varphi(\pi / 4)-\varphi_{0}\right|=0$, and the corresponding values of $\log _{10}\left|\varphi(\pi / 4)-\varphi_{0}\right|$.


Figure 4.13: Dependence of the parameter $\tilde{\lambda}_{\theta}$ on parameter $\tilde{\mu}$ obtained from Table 4.3.

Using these values of the couple ( $\tilde{\mu}, \tilde{\lambda}_{\theta}$ ) shown in Figure 4.13, we are now in position to solve the differential equation (4.151a), to determine the value of $t_{0}$ when $\varphi\left(t_{0}\right)=\varphi_{0}$, and thus to obtain the optimal time $T=2 t_{0}$, and the resulting time area of the transient population in the excited state (4.8) (4.136b): $A=\int_{t_{i}}^{t_{f}} d t x_{2}(t)^{2}=2 \int_{0}^{t_{0}} d t \sin ^{2} \varphi(t)$. We show the corresponding data in Table 4.4, from which one can conclude that the optimal control time $T$ decreases for increasing values of $A$ with diminishing rates and gradually flattens out for large $A$ (see Figure 4.14).

| $\tilde{\mu}$ | $\tilde{\lambda}_{\theta}$ | $\varphi_{0}$ | A <br> (in units of $1 / u_{0}$ ) | $\begin{gathered} T \\ \text { (in units } \\ \text { of } 1 / u_{0} \text { ) } \end{gathered}$ | $A / T$ | $\mathcal{A}$ | $\begin{gathered} \mathcal{E} \\ \text { (in units } \\ \text { of } \hbar / T \text { ) } \end{gathered}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| 0.5 | 0.25879 | 1.1527 | 1.1177 | 2.74770 | 0.4068 | 2.75 | 7.54 |
| 0 | 0.57735 | 1.0472 | 1.0203 | 2.72070 | 0.3750 | 2.72 | 7.40 |
| -0.5 | 0.90476 | 0.9783 | 0.9612 | 2.73455 | 0.3515 | 2.73 | 7.48 |
| -1 | 1.23456 | 0.9248 | 0.9176 | 2.76679 | 0.3317 | 2.77 | 7.65 |


| -1.5 | 1.56434 | 0.8795 | 0.8821 | 2.81091 | 0.3138 | 2.81 | 7.90 |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| -2 | 1.89263 | 0.8389 | 0.8513 | 2.86467 | 0.2972 | 2.86 | 8.21 |
| -2.5 | 2.21825 | 0.8012 | 0.8234 | 2.92745 | 0.2813 | 2.93 | 8.57 |
| -3 | 2.53997 | 0.7652 | 0.7972 | 2.99944 | 0.2658 | 3.00 | 9.00 |
| -3.5 | 2.85642 | 0.7301 | 0.7720 | 3.08129 | 0.2505 | 3.08 | 9.49 |
| -4 | 3.16595 | 0.6955 | 0.7473 | 3.17385 | 0.2355 | 3.17 | 10.07 |
| -4.5 | 3.46665 | 0.6609 | 0.7228 | 3.27788 | 0.2205 | 3.28 | 10.74 |
| -5 | 3.75639 | 0.6266 | 0.6984 | 3.39383 | 0.2058 | 3.39 | 11.52 |
| -5.5 | 4.03322 | 0.5926 | 0.6742 | 3.52108 | 0.1915 | 3.52 | 12.40 |
| -6 | 4.29565 | 0.5597 | 0.6504 | 3.65797 | 0.1778 | 3.66 | 13.38 |
| -6.5 | 4.543113 | 0.5283 | 0.6273 | 3.80182 | 0.1650 | 3.80 | 14.45 |
| -7 | 4.776073 | 0.4992 | 0.6055 | 3.94941 | 0.1533 | 3.95 | 15.60 |
| -7.5 | 4.995772 | 0.4727 | 0.5850 | 4.09787 | 0.1428 | 4.10 | 16.79 |
| -8 | 5.203814 | 0.4489 | 0.5660 | 4.24493 | 0.1333 | 4.24 | 18.02 |
| -8.5 | 5.401802 | 0.4277 | 0.5485 | 4.38933 | 0.1250 | 4.39 | 19.27 |
| -9 | 5.591159 | 0.4088 | 0.5323 | 4.52959 | 0.1175 | 4.53 | 20.52 |
| -9.5 | 5.773058 | 0.3920 | 0.5175 | 4.66656 | 0.1109 | 4.67 | 21.78 |
| -10 | 5.948450 | 0.3770 | 0.5037 | 4.79884 | 0.1050 | 4.80 | 23.03 |
| -10.5 | 6.1180957 | 0.3635 | 0.4913 | 4.92992 | 0.0997 | 4.93 | 24.30 |
| -11 | 6.2826117 | 0.3514 | 0.4794 | 5.05505 | 0.0948 | 5.06 | 25.55 |
| -11.5 | 6.4425007 | 0.3405 | 0.4684 | 5.17677 | 0.0905 | 5.18 | 26.80 |
| -12 | 6.5981805 | 0.3305 | 0.4582 | 5.29634 | 0.0865 | 5.30 | 28.05 |
| -12.5 | 6.75000296 | 0.3214 | 0.4488 | 5.41477 | 0.0829 | 5.41 | 29.32 |
| -13 | 6.89826843 | 0.3130 | 0.4397 | 5.52782 | 0.0795 | 5.53 | 30.56 |
| -13.5 | 7.04323635 | 0.3053 | 0.4311 | 5.63863 | 0.0765 | 5.64 | 31.79 |
| -14 | 7.185133286 | 0.2981 | 0.4235 | 5.75250 | 0.0736 | 5.75 | 33.09 |
| -14.5 | 7.324158901 | 0.2915 | 0.4160 | 5.85996 | 0.0710 | 5.86 | 34.34 |
| -15 | 7.460490528 | 0.2853 | 0.4085 | 5.96127 | 0.0685 | 5.96 | 35.54 |
| -15.5 | 7.5942867762 | 0.2795 | 0.4020 | 6.06896 | 0.0662 | 6.07 | 36.83 |
| -16 | 7.7256903690 | 0.2740 | 0.3957 | 6.17221 | 0.0641 | 6.17 | 38.10 |
| -16.5 | 7.8548304148 | 0.2689 | 0.3892 | 6.26802 | 0.0621 | 6.27 | 39.29 |
| -17 | 7.98182426212 | 0.2640 | 0.3836 | 6.36988 | 0.0602 | 6.37 | 40.58 |
| -17.5 | 8.10677902312 | 0.2594 | 0.3779 | 6.46565 | 0.0584 | 6.47 | 41.80 |
| -18 | 8.22979283925 | 0.2551 | 0.3730 | 6.56874 | 0.0568 | 6.57 | 43.15 |
| -18.5 | 8.35095594416 | 0.2509 | 0.3671 | 6.65174 | 0.0552 | 6.65 | 44.25 |
| -19 | 8.47035156274 | 0.2470 | 0.3622 | 6.74343 | 0.0537 | 6.74 | 45.47 |
|  |  |  |  |  |  |  |  |
| -1 |  |  |  |  |  |  |  |


| -19.5 | 8.588056677662 | 0.2432 | 0.3577 | 6.83913 | 0.0523 | 6.84 | 46.77 |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| -20 | 8.704142687429 | 0.2397 | 0.3531 | 6.92962 | 0.0510 | 6.93 | 48.02 |
| -20.5 | 8.8186759747537 | 0.2362 | 0.3486 | 7.01618 | 0.0497 | 7.02 | 49.23 |
| -21 | 8.9317184002888 | 0.2330 | 0.3446 | 7.10736 | 0.0485 | 7.11 | 50.51 |
| -21.5 | 9.0433277337173 | 0.2298 | 0.3406 | 7.19646 | 0.0473 | 7.20 | 51.79 |
| -22 | 9.1535580319427 | 0.2268 | 0.3368 | 7.28429 | 0.0462 | 7.28 | 53.06 |
| -22.5 | 9.262459972324599 | 0.2239 | 0.3325 | 7.35882 | 0.0452 | 7.36 | 54.15 |
| -23 | 9.370081147493655 | 0.2211 | 0.3264 | 7.39236 | 0.0442 | 7.39 | 54.65 |
| -23.5 | 9.476466327158352 | 0.2184 | 0.3234 | 7.48637 | 0.0432 | 7.49 | 56.05 |
| -24 | 9.581657691415900 | 0.2159 | 0.3225 | 7.62303 | 0.0423 | 7.62 | 58.11 |
| -24.5 | 9.685695039350745 | 0.2134 | 0.3190 | 7.70057 | 0.0414 | 7.70 | 59.30 |

Table 4.4: Resulting values of $\varphi_{0}$ (i.e. $\varphi(T / 2)$ ), the time area of the transient population in the excited state $A$ and the optimal control time $T$ for various couples of $\left(\tilde{\mu}, \tilde{\lambda}_{\theta}\right)$. The pulse energy $\mathcal{E}$, Eq. (4.7), corresponds to the value given for optimal time but in units of $u_{0}$. The pulse area $\mathcal{A}=\int_{0}^{T} d s \sqrt{u_{p}^{2}+u_{s}^{2}}$ is also given (which corresponds in fact to the value of $\left.u_{0} T\right)$.


Figure 4.14: Dependence of the optimal control time $T$ on the area $A$ obtained from Table 4.4.

We observe from Table 4.4 that, for the unconstrained case, i.e. $\tilde{\mu}=0$, the determined time area is $A \simeq 1.0203 / u_{0}$ while the optimal time is $T=2.72070 / u_{0}$, which recovers the unconstrained result obtained in the energy-optimal dissipative STIREP (see Table 4.2)(i.e. Eq. (4.84)): $A=\frac{3 T}{8} \simeq 1.0203 / u_{0}$, and $\varphi_{0}=1.0472$. Comparing the constrained cases with the ones of energy-optimal dissipative STIREP (see Table 4.2), for instance, when $A / T=0.05$, the energy for energy-optimal STIREP is $\mathcal{E}_{\text {energy-optimal }} \simeq$ $40 \frac{\hbar}{T}$, markedly smaller than time-optimal dissipative STIREP $\mathcal{E}_{\text {time-optimal }} \simeq 49 \frac{\hbar}{T}$; while the control time for energy-optimal STIREP is $T_{\text {energy-optimal }} \simeq 14 / u_{0}$, nearly twice larger than time-optimal dissipative STIREP $T_{\text {time-optimal }} \simeq 7 / u_{0}$. We also notice that the pulse area for energy-optimal STIREP is smaller than time-optimal STIREP. Still take $A / T=0.05$ as an example, $\mathcal{A}_{\text {energy-optimal }} \simeq \pi$, almost one-half of $\mathcal{A}_{\text {time-optimal }} \simeq 7$.

Furthermore, such values of $\tilde{\mu}, \tilde{\lambda}_{\theta}$, and the corresponding obtained optimal control time $T$ shown in Table 4.4 lead to the the design of the pulses. Details of the analyses are presented below in Subsection 4.3.3.

### 4.3.3 Derivation of the pulses and the dynamics

When we get pairs of $\left(\tilde{\mu}, \tilde{\lambda}_{\theta}\right)$ with specific values, $\varphi$ thus can be obtained numerically by solving the differential equation (4.151a):

$$
\begin{equation*}
\dot{\varphi}= \pm \frac{\sqrt{\left(1-\tilde{\mu} \sin ^{2} \varphi\right)^{2}-\tilde{\lambda}_{\theta}^{2} \tan ^{2} \varphi}}{1-\tilde{\mu} \sin ^{2} \varphi} \tag{4.177}
\end{equation*}
$$

and then $\lambda_{\varphi}$ is acquirable from (4.148):

$$
\begin{equation*}
\lambda_{\varphi}= \pm \sqrt{\left(h-\mu \sin ^{2} \varphi\right)^{2}-\lambda_{\theta}^{2} \tan ^{2} \varphi} \tag{4.178}
\end{equation*}
$$

Based on Eq.(4.139), the original controls can be written as

$$
\begin{align*}
& \tilde{u}_{p}=\tilde{v}_{p} \cos \theta-\tilde{v}_{s} \sin \theta  \tag{4.179a}\\
& \tilde{u}_{s}=-\tilde{v}_{p} \sin \theta-\tilde{v}_{s} \cos \theta \tag{4.179b}
\end{align*}
$$

where $\tilde{v}_{p}$ and $\tilde{v}_{s}$ are given by (4.144):

$$
\begin{align*}
\tilde{v}_{p} & =\frac{\lambda_{\varphi}}{R}  \tag{4.180a}\\
\tilde{v}_{s} & =-\frac{\lambda_{\theta} \tan \varphi}{R} \tag{4.180b}
\end{align*}
$$

with

$$
\begin{equation*}
R=\sqrt{\lambda_{\varphi}^{2}+\lambda_{\theta}^{2} \tan ^{2} \varphi} \tag{4.181}
\end{equation*}
$$

(see Eq. (4.145)), and $\theta$ can be derived numerically from the transformation of the differential equation (4.151b):

$$
\begin{equation*}
\frac{d \theta}{d \varphi}= \pm \frac{\tilde{\lambda}_{\theta} \tan ^{2} \varphi}{\sqrt{\left(1-\tilde{\mu} \sin ^{2} \varphi\right)^{2}-\tilde{\lambda}_{\theta}^{2} \tan ^{2} \varphi}} \tag{4.182}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
\theta= \pm \int_{\varphi_{i}}^{\varphi_{f}} \frac{\tilde{\lambda}_{\theta} \tan ^{2} \varphi}{\sqrt{\left(1-\tilde{\mu} \sin ^{2} \varphi\right)^{2}-\tilde{\lambda}_{\theta}^{2} \tan ^{2} \varphi}} d \varphi \tag{4.183}
\end{equation*}
$$

From the values $\tilde{\mu}, \tilde{\lambda}_{\theta}$ and the optimal time $T$ shown in Table 4.4, one can derive the controls and the resulting numerics of the populations. Figures 4.15-4.19 show the parameters $\varphi(t)$ and $\theta(t)$ and the dynamics for instance for five couples of $\left(\tilde{\mu}, \tilde{\lambda}_{\theta}\right)$ and the corresponding obtained $T$, respectively. Figure 4.20-4.27 compares controls for some values of $\left(\tilde{\mu}, \tilde{\lambda}_{\theta}\right)$ and $T$ of Table 4.4. We observe that as the optimal time increases (i.e. $\tilde{\mu}$ becomes larger in negative), the pump pulse decreases sharply at early times and symmetrically the Stokes pulse increases sharply at late times, and then increases to the maximum at almost the end of the control duration, which features a counterintuitive sequence. The projection of the dynamics onto a dark state defined with the actual angle $\theta$ gets closer to one for a smaller admissible loss. We can notice that this projection is in fact 1 minus the population in state 2 .


Figure 4.15: Time-dependence of the parameters $\varphi$ (upper frame, solid cyan line) and $\theta$ (upper frame, solid magenta line), the control pulses $u_{p}$ (upper middle frame, solid red line) and $u_{s}$ (upper middle frame, solid blue line) (in units of $u_{0}$ ), the projection (in absolute value squared) of the dynamics onto the dark state defined with the actual $\theta$ (lower middle frame), and populations (lower frame) for $\tilde{\mu}=0$ and the resulting optimal time $T=2.72 / u_{0}$ shown in Table 4.4 corresponding to unconstrained optimal pulses.


Figure 4.16: Same as Figure 4.15, but for $\tilde{\mu}=-5$.
4.3. Optimal control with respect to time constrained by a given admissible loss: time-optimal dissipative STIREP


Figure 4.17: Same as Figure 4.15 , but for $\tilde{\mu}=-10.5$.


Figure 4.18: Same as Figure 4.15 , but for $\tilde{\mu}=-20.5$.
4.3. Optimal control with respect to time constrained by a given admissible loss: time-optimal dissipative STIREP


Figure 4.19: Same as Figure 4.15, but for $\tilde{\mu}=-24$.


Figure 4.20: Comparison of the controls (in units of $u_{0}$ ) for various couples of $\left(\tilde{\mu}, \tilde{\lambda}_{\theta}\right)$ and the corresponding optimal time $T$ from Table 4.4.


Figure 4.21: Same as Figure 4.20 but for larger negative values of $\tilde{\mu}$ and the corresopnding $\tilde{\lambda}_{\theta}$ and optimal time $T$ from Table 4.4.
4.3. Optimal control with respect to time constrained by a given admissible loss: time-optimal dissipative STIREP


Figure 4.22: Controls (in units of $u_{0}$ ) for $\tilde{\mu}=-11, \tilde{\lambda}_{\theta}=6.2826117$ and optimal time $T=5.06 / u_{0}$ from Table 4.4.


Figure 4.23: Controls (in units of $u_{0}$ ) for $\tilde{\mu}=-15, \tilde{\lambda}_{\theta}=7.460490528$ and optimal time $T=5.96 / u_{0}$ from Table 4.4.


Figure 4.24: Controls (in units of $u_{0}$ ) for $\tilde{\mu}=-18, \tilde{\lambda}_{\theta}=8.22979283925$ and optimal time $T=6.57 / u_{0}$ from Table 4.4.


Figure 4.25: Controls (in units of $u_{0}$ ) for $\tilde{\mu}=-19, \tilde{\lambda}_{\theta}=8.47035156274$ and optimal time $T=6.74 / u_{0}$ from Table 4.4.
4.3. Optimal control with respect to time constrained by a given admissible loss: time-optimal dissipative STIREP


Figure 4.26: Controls (in units of $u_{0}$ ) for $\tilde{\mu}=-22, \tilde{\lambda}_{\theta}=9.1535580319427$ and optimal time $T=7.28 / u_{0}$ from Table 4.4.


Figure 4.27: Controls (in units of $u_{0}$ ) for $\tilde{\mu}=-24, \tilde{\lambda}_{\theta}=9.581657691415900$ and optimal time $T=7.62 / u_{0}$ from Table 4.4.

### 4.4 Discussion

In this chapter, we derive energy- (Section 4.2) and time-minimum (Section 4.3) optimizations under the constraint of a given admissible loss leading to exact state-tostate transfer of three-level $\Lambda$-type quantum systems. The analytical expression of the external control fields is also obtained. We notice that the resulting control fields in such two cases are quite different. More specifically, in the case of energy-optimal dissipative STIREP, the pulse amplitude of the optimal result increases with the decrease of the given loss and features an overlapping sequence once the loss decreases to a certain value; in the case of time-optimal dissipative STIREP, the control time increases with the decrease of the given loss and characterizes a counterintuitive sequence once the loss decreases to a certain value. The energy-optimal strategy operates in a fast way (necessitating a relatively strong field) and far from a dark state. On the other hand, the time-optimal strategy operates relatively close to the dark state in the limit of a low admissible loss and features some similarities with adiabatic passage except the very beginning and the very end of the process, which are strongly non-adiabatic.


Figure 4.28: Pulse amplitudes for the three examples (i), (ii), (iii) described in the text. For (ii) and (iii), we have only plotted $u_{0}$.

The process duration, peak pulse amplitude, energy, and area, for several given admissible losses are put down in Tables 4.2 and 4.4. One can compare a few values in order to exhibit the different strategies: for instance, (i) for the given admissible loss $A=0.05 / T$ in Table 4.2, we have $A \approx 0.7 / u_{\max }$, the time of operation $T \approx 14 / u_{\max }$, the energy $\mathcal{E}=40 \hbar / T \approx 2.8 u_{\max }$ and the pulse area $\mathcal{A}=\pi$. (ii) In Table 4.4, the same loss (for the same peak amplitude $u_{0}$ ) is roughly obtained for $\tilde{\mu}=-5$ : $A \approx 0.7 / u_{0}$, $T \approx 3.4 / u_{0}, \mathcal{E} \approx 11.5 \hbar / T \approx 3.4 u_{0}$ and $\mathcal{A}=3.4$, which shows a much smaller (roughly four times smaller) time of operation but a larger pulse area and energy. On the other hand, in Table 4.4, (iii) the same loss $A=0.05 / T$ as in (i) (for the same duration $T)$ is roughly obtained for $\tilde{\mu}=-20.5$, which leads to a larger energy $\mathcal{E}=49 \hbar / T$ and a larger pulse area $\mathcal{A}=7$, but to a (twice) smaller peak pulse amplitude $u_{0} \approx 7 / T$. We show the dependence of the pulse amplitude on the duration corresponding to such three examples in Figure 4.28. The energy-minimization strategy can thus achieve the transfer with a given loss in a fast way in the sense that the pulse area is small, but for a large peak pulse amplitude. On the other hand, the time-minimization strategy can achieve it with a weaker pulse amplitude, but for a larger pulse area (and energy).


Figure 4.29: Final population transfer as a function of the deviation $\varepsilon$ for energy-optimal dissipative STIREP with respect to various time area of the transient population in the excited state $A=0.375 T$ (unconstrained case) (solid blue line), $A=0.1 T$ (solid red line) and $A=0.05 T$ (dashed green line), respectively.


Figure 4.30: Comparison of final population transfer as a function of the deviation $\varepsilon$ between time-optimal dissipative STIREP (solid lines) and STIRAP (dashed lines) with respect to various time area of the transient population in the excited state $A=0.375 T$ (unconstrained case) (blue lines), $A=0.1 T$ (red lines) and $A=0.05 T$ (green lines), respectively.

The comparison of their robustness between the two cases, for instance, considering typically three sets of same dissipation, is next shown in Figures 4.29 and 4.30 (solid lines). The notation $\varepsilon$ represents the relative deviation on the control field amplitude. We observe that, comparing with the energy-optimal dissipative STIREP, the controls determined from time-optimal dissipative STIREP are more robust when the given loss (i.e. time area of the transient population in the excited state) is low. The weak robustness for the energy-minimization process can be understood by the fact that this procedure finds the counterpart of the two-state $\pi$-pulse transfer to the three-state system with a large amplitude and a short time in order to respect the admissible loss.

We also compare the robustness for STIRAP with time-optimal dissipative STIREP, whose deviation profile is depicted in Figure 4.30 (dashed lines). It shows that the final population transfer as a function of $\varepsilon$ for STIRAP is flatter with respect to relatively
small loss. But the transfer is more precise for time-optimal dissipative STIREP when $\varepsilon=0$ especially for relatively small loss.

The corresponding dynamics profiles for STIRAP are shown in Figures 4.31, 4.32 and 4.33. Comparing with the pulses we derived in time-optimal dissipative STIREP which feature the same time area (Figures 4.15, 4.17 and 4.18), respectively, we notice that the duration of STIRAP is observably longer than time-optimal dissipative STIREP. The pulse area for STIRAP gives $\mathcal{A}_{\text {STIRAP }} \simeq 4$ for $A=0.375 T$, $\mathcal{A}_{\text {STIRAP }} \simeq 10$ for $A=0.1 T$ and $\mathcal{A}_{\text {STIRAP }} \simeq 17$ for $A=0.05 T$, noticeably larger than time-optimal dissipative STIREP (see Table 4.4) $\mathcal{A}_{\text {time-optimal }} \simeq 3$ for $A=0.375 T, \mathcal{A}_{\text {time-optimal }} \simeq 5$ for $A=0.1 T$ and $\mathcal{A}_{\text {time-optimal }} \simeq 7$ for $A=0.05 T$.


Figure 4.31: Stokes $u_{s}$ (upper frame, solid red line) and Pump $u_{p}$ (upper frame, solid black line) (in units of $1 / T$ ) for STIRAP, the projection (in absolute value squared) of the dynamics onto the dark state defined with the actual $\theta$ (middle frame) and the resulting populations with $\psi(t)$ the state solution (lower frame) in a situation of time area of the transient population in the excited state $A=0.375 T$.


Figure 4.32: Same as Figure 4.31 but for $A=0.1 T$.


Figure 4.33: Same as Figure 4.31 but for $A=0.05 T$.

## Optimal robust quantum control by inverse optimization: robust inverse optimization (RIO) method

## Contents

5.1 The model and principles of inverse engineering ..... 132
5.2 The single-shot shaped pulse method ..... 134
5.3 Robust population transfer - Figure of merit ..... 136
5.4 The unconstrained optimization problem ..... 138
5.5 Application of RIO for robustness with respect to field inho- mogeneities $(\delta=0)$ ..... 140
5.5.1 Complete population transfer ..... 140
5.5.2 Robust trajectory optimal with respect to pulse energy or time ..... 147
5.5.3 Partial population transfer ..... 150
5.6 Discussion ..... 153

In this chapter, we propose an alternative method of geometric optimization based on inverse engineering:

- We apply in a first stage the Euler-Lagrange optimization [57] constrained by robustness integrals and by the boundaries ensuring exact fidelity in the dynamical variable space without invoking the dynamical equations. More precisely, we formulate the problem with trajectories that exactly connect the initial state to the defined target and that are constrained by the robustness via simple integrals. The constrained Euler-Lagrange optimization leads to a robust geodesic, i.e. the optimal trajectory with respect to a given cost under the constraint of robustness.
- In a second stage, we derive the control fields from the geodesic using the dynamical equation, i.e. the time-dependent Schrödinger equation for the present quantum control problem, formulated in an inverted way, in which we express the Hamiltonian elements (the controls) from the dynamical variables.

The method is referred to as robust inverse optimization (RIO). It is applied to determine complete and partial transfers that feature both properties in terms of control pulse area (chosen as the cost): optimality and robustness.

Section 5.1 introduces the model with the dynamical variable space we study. The robustness is formulated by the single-shot shaped pulse method (SSSP) in Section 5.2. Section 5.3 focuses on the figure of merit for robust population transfer through integrals. In Section 5.4, we rederive the unconstrained (non-robust) optimization solution reformulated with the Euler-Lagrange optimization. Section 5.5 presents technical computations in detail showing the derivation of the optimal robust solutions for complete and partial population transfer with respect to field inhomogeneities. The discussion in Section 5.6 gives the conclusion of this chapter and some suggestions for further applications.

### 5.1 The model and principles of inverse engineering

We consider the Hamiltonian $H_{\lambda}=H_{0}+\lambda V$ (in units such that $\hbar=1$ ), where

$$
H_{0}=\frac{1}{2}\left[\begin{array}{cc}
-\Delta & \Omega  \tag{5.1}\\
\Omega & \Delta
\end{array}\right]
$$

models the qubit $\{|0\rangle,|1\rangle\}$ with the control parameters: the Rabi frequency $\Omega \equiv \Omega(t)$ (considered positive for simplicity and without loss of generality) and the detuning $\Delta \equiv \Delta(t)$, and $\lambda$ gathers unknown (time-independent) parameters representing the systematic errors in the description of the model. This can be due to an imperfect knowledge of the quantum system or of the interacting pulse (such as the positioning of the quantum system with respect to the interacting pulse, field inhomogeneities, homogeneous or inhomogeneous broadening, ...). We consider the errors that can be modeled by static deviation parameters. An error (i) with respect to the pulse amplitude (or area) is modeled with $V=\Omega \sigma_{x} / 2$ and $\lambda=\alpha$ corresponds to the relative deviation of the pulse amplitude (field inhomogeneties); (ii) with respect to the detuning is modeled with $V=-\sigma_{z} / 2$ and $\lambda=\delta$ corresponds to the absolute deviation of the detuning.

The solution of the TDSE: $i \hbar \frac{\partial}{\partial t}\left|\phi_{0}(t)\right\rangle=H_{0}\left|\phi_{0}(t)\right\rangle$ is conveniently parameterized with two angles $\theta \equiv \theta(t) \in[0, \pi]$, the internal (or relative) phase $\varphi \equiv \varphi(t) \in[-\pi, \pi]$ and a global phase $\gamma \equiv \gamma(t) \in[0,2 \pi]$ as

$$
\left|\phi_{0}(t)\right\rangle=\left[\begin{array}{c}
\cos \frac{\theta}{2} e^{i \varphi / 2}  \tag{5.2}\\
\sin \frac{\theta}{2} e^{-i \varphi / 2}
\end{array}\right] e^{-i \gamma / 2}
$$

Inserting it into the TDSE, we obtain

$$
\begin{align*}
\dot{\theta} & =\Omega \sin \varphi  \tag{5.3a}\\
\dot{\varphi} & =\Delta+\Omega \cos \varphi \cot \theta  \tag{5.3b}\\
\dot{\gamma} & =\Omega \frac{\cos \varphi}{\sin \theta}=\dot{\theta} \frac{\cot \varphi}{\sin \theta} \tag{5.3c}
\end{align*}
$$

where the dot represents the derivation with respect to time $t$. The inverse-engineering method consists in determining the Hamiltonian elements (the controls) from the dynamical variables by inverting the TDSE: $H_{0}=i \hbar\left(\frac{\partial}{\partial t} U_{0}\left(t, t_{i}\right)\right) U_{0}^{\dagger}\left(t, t_{i}\right)$, i.e. from inversion of Eqs. (5.3):

$$
\begin{align*}
\Delta & =\dot{\varphi}-\dot{\gamma} \cos \theta  \tag{5.4a}\\
\Omega & =\sqrt{\dot{\theta}^{2}+\dot{\gamma}^{2} \sin ^{2} \theta}  \tag{5.4~b}\\
& =|\dot{\theta}| \sqrt{1+\left(\frac{d \widetilde{\gamma}}{d \theta}\right)^{2} \sin ^{2} \theta}  \tag{5.4c}\\
& =|\dot{\gamma}| \sqrt{\left(\frac{d \widetilde{\theta}}{d \gamma}\right)^{2}+\sin ^{2} \widetilde{\theta}} \tag{5.4~d}
\end{align*}
$$

In Eq. (5.4c), we have assumed that one can write $\gamma(t)$ as a function of $\theta: \widetilde{\gamma}(\theta) \equiv \gamma(t)$. In Eq. (5.4d), we have assumed on the other hand that one can write $\theta(t)$ as a function of $\gamma: \widetilde{\theta}(\gamma) \equiv \theta(t)$. We note that the pulse area from the initial $t_{i}$ to the final $t_{f}$ times (denoting $\theta_{i} \equiv \theta\left(t_{i}\right), \theta_{f} \equiv \theta\left(t_{f}\right)$ and assuming a monotonic $\theta(t)$ such that $\dot{\theta}>0$ )

$$
\begin{equation*}
\int_{t_{i}}^{t_{f}} d t \Omega(t)=\int_{\theta_{i}}^{\theta_{f}} d \theta \sqrt{1+\left(\frac{d \widetilde{\gamma}}{d \theta}\right)^{2} \sin ^{2} \theta} \tag{5.5}
\end{equation*}
$$

does not depend on the time-dependence of $\theta(t)$, but only on the derivative of the function $\widetilde{\gamma}(\theta)$. Alternatively, one can write the pulse area (denoting $\gamma_{i} \equiv \gamma\left(t_{i}\right), \gamma_{f} \equiv$ $\gamma\left(t_{f}\right)$ and assuming a monotonic $\gamma(t)$ such that $\left.\dot{\gamma}>0\right)$ as

$$
\begin{equation*}
\int_{t_{i}}^{t_{f}} d t \Omega(t)=\int_{\gamma_{i}}^{\gamma_{f}} d \gamma \sqrt{\left(\frac{d \widetilde{\theta}}{d \gamma}\right)^{2}+\sin ^{2} \widetilde{\theta}} \tag{5.6}
\end{equation*}
$$

which does not depend on the time-dependence of $\gamma(t)$, but only on $\widetilde{\theta}(\gamma)$ and its derivative.
In general we will be led to consider multiple functions $\widetilde{\gamma}_{j}(\theta)$ or $\widetilde{\theta}_{j}(\gamma)$ (see (5.24)), each depending on the time interval, in order to get the optimal solution.

We denote $\left|\phi_{\lambda}(t)\right\rangle$ the state of the complete dynamics, solution of the TDSE $i \hbar \frac{\partial}{\partial t}\left|\phi_{\lambda}(t)\right\rangle=H_{\lambda}\left|\phi_{\lambda}(t)\right\rangle$.

We will see that the integrals characterizing the robustness will depend on functions of $\theta$ and $\gamma$ (see Eqs. (5.9), (5.10) and (5.11)), as well as the cost integral of optimization (such as the pulse area). We will then consider the two dynamical variables $\theta(t)$ and $\gamma(t)$ providing a geometric representation of the problem, and the third variable $\varphi(t)$ is given by $(5.3 \mathrm{c}), \cot \varphi=\dot{\gamma} \sin \theta / \dot{\theta}$, from which we obtain

$$
\begin{equation*}
\dot{\varphi}=\frac{\ddot{\theta} \dot{\gamma} \sin \theta-\ddot{\gamma} \dot{\theta} \sin \theta-\dot{\gamma} \dot{\theta}^{2} \cos \theta}{\dot{\theta}^{2}+\dot{\gamma}^{2} \sin ^{2} \theta} \tag{5.7}
\end{equation*}
$$

### 5.2 The single-shot shaped pulse method

The SSSP method, defined in [35], can be summarized as follows (where $\lambda \equiv(\alpha, \delta)$ ). The perturbative expansion of $\phi_{\alpha, \delta}\left(t_{f}\right)$ with respect to $\alpha$ and $\delta$ reads

$$
\begin{equation*}
\left\langle\phi_{T} \mid \phi_{\alpha, \delta}\left(t_{f}\right)\right\rangle=1+O_{1}+O_{2}+O_{3}+\cdots, \tag{5.8}
\end{equation*}
$$

where $O_{n}$ denotes the term of total order $n: O_{n} \equiv O\left(\lambda^{n}\right)$ and $\left|\phi_{T}\right\rangle$ the target state. The first four terms read

$$
\begin{align*}
O_{1}= & -i \int_{t_{i}}^{t_{f}} e(t) d t,  \tag{5.9a}\\
O_{2}= & (-i)^{2} \int_{t_{i}}^{t_{f}} d t \int_{t_{i}}^{t} d t^{\prime}\left[e(t) e\left(t^{\prime}\right)+f(t) \bar{f}\left(t^{\prime}\right)\right],  \tag{5.9b}\\
O_{3}= & (-i)^{3} \int_{t_{i}}^{t_{f}} d t \int_{t_{i}}^{t} d t^{\prime} \int_{t_{i}}^{t^{\prime}} d t^{\prime \prime}\left[e(t) e\left(t^{\prime}\right) e\left(t^{\prime \prime}\right)\right. \\
& \left.+e(t) f\left(t^{\prime}\right) \bar{f}\left(t^{\prime \prime}\right)+f(t) \bar{f}\left(t^{\prime}\right) e\left(t^{\prime \prime}\right)-f(t) e\left(t^{\prime}\right) \bar{f}\left(t^{\prime \prime}\right)\right],  \tag{5.9c}\\
O_{4}= & (-i)^{4} \int_{t_{i}}^{t_{f}} d t \int_{t_{i}}^{t} d t^{\prime} \int_{t_{i}}^{t^{\prime}} d t^{\prime \prime} \int_{t_{i}}^{t^{\prime \prime}} d t^{\prime \prime \prime} \\
& {\left[e(t) e\left(t^{\prime}\right) e\left(t^{\prime \prime}\right) e\left(t^{\prime \prime \prime}\right)+e(t) e\left(t^{\prime}\right) f\left(t^{\prime \prime}\right) \bar{f}\left(t^{\prime \prime \prime}\right)\right.} \\
& +e(t) f\left(t^{\prime}\right) \bar{f}\left(t^{\prime \prime}\right) e\left(t^{\prime \prime \prime}\right)-e(t) f\left(t^{\prime}\right) e\left(t^{\prime \prime}\right) \bar{f}\left(t^{\prime \prime \prime}\right) \\
& +f(t) \bar{f}\left(t^{\prime}\right) e\left(t^{\prime \prime}\right) e\left(t^{\prime \prime \prime}\right)+f(t) \bar{f}\left(t^{\prime}\right) f\left(t^{\prime \prime}\right) \bar{f}\left(t^{\prime \prime \prime}\right) \\
& \left.-f(t) e\left(t^{\prime}\right) \bar{f}\left(t^{\prime \prime}\right) e\left(t^{\prime \prime \prime}\right)+f(t) e\left(t^{\prime}\right) e\left(t^{\prime \prime}\right) \bar{f}\left(t^{\prime \prime \prime}\right)\right] \tag{5.9d}
\end{align*}
$$

with

$$
\begin{gather*}
e \equiv\left\langle\phi_{0}(t)\right| V(t)\left|\phi_{0}(t)\right\rangle=-\frac{1}{2}\left(\delta \cos \theta-\alpha \dot{\gamma} \sin ^{2} \theta\right)  \tag{5.10}\\
f \equiv\left\langle\phi_{0}(t)\right| V(t)\left|\phi_{\perp}(t)\right\rangle=\frac{1}{2}\left[\delta \sin \theta+\alpha\left(\frac{1}{2} \dot{\gamma} \sin 2 \theta-i \dot{\theta}\right)\right] e^{i \gamma} \tag{5.11}
\end{gather*}
$$

and the orthogonal solution of the TDSE:

$$
\left|\phi_{\perp}(t)\right\rangle=\left[\begin{array}{c}
e^{i \varphi / 2} \sin (\theta / 2)  \tag{5.12}\\
-e^{-i \varphi / 2} \cos (\theta / 2)
\end{array}\right] e^{i \gamma / 2}
$$

such that $\left\langle\phi_{\perp}(t) \mid \phi_{0}(t)\right\rangle=0$. The other terms can be determined from a symbolic diagram [35], see Figure 5.1.




Figure 5.1: Symbolic path diagrams giving the construction of the $O_{n}$ integrals. The symbol $e$ stands for $e(t), e^{\prime}$ for $e\left(t^{\prime}\right)$, and so on. For instance, for $n=3$, the diagram features four paths. Its extension for larger $n$ is direct.

We denote at a certain order $n$

$$
\begin{equation*}
\left\langle\phi_{T} \mid \phi_{\alpha, \delta}\left(t_{f}\right)\right\rangle_{n}=1+O_{1}+O_{2}+\cdots+O_{n} \tag{5.13}
\end{equation*}
$$

The integral $O_{2}$ can be simplified using the property

$$
\begin{equation*}
\int_{\tau}^{T} d t \int_{\tau}^{t} d t^{\prime}\left[a(t) b\left(t^{\prime}\right)+a\left(t^{\prime}\right) b(t)\right]=\int_{\tau}^{T} a(t) d t \int_{\tau}^{T} b(t) d t \tag{5.14}
\end{equation*}
$$

from

$$
\begin{equation*}
\int_{\tau}^{T} v d u+\int_{\tau}^{T} u d v=[u v]_{\tau}^{T} \tag{5.15}
\end{equation*}
$$

where

$$
\begin{array}{ll}
u(t)=\int_{\tau}^{t} a\left(t^{\prime}\right) d t^{\prime}, & d u=a(t) d t \\
v(t)=\int_{\tau}^{t} b\left(t^{\prime}\right) d t^{\prime}, & d v=b(t) d t \tag{5.16b}
\end{array}
$$

For $a=b \equiv e$, we have

$$
\begin{equation*}
\int_{t_{i}}^{t_{f}} d t \int_{t_{i}}^{t} d t^{\prime}\left[e(t) e\left(t^{\prime}\right)+e\left(t^{\prime}\right) e(t)\right]=2 \int_{t_{i}}^{t_{f}} d t \int_{t_{i}}^{t} d t^{\prime} e(t) e\left(t^{\prime}\right)=\left[\int_{t_{i}}^{t_{f}} e(t) d t\right]^{2} \tag{5.17}
\end{equation*}
$$

For $a \equiv f$ and $b \equiv \bar{f}$, we have

$$
\begin{equation*}
\int_{t_{i}}^{t_{f}} d t \int_{t_{i}}^{t} d t^{\prime}\left[f(t) \bar{f}\left(t^{\prime}\right)+f\left(t^{\prime}\right) \bar{f}(t)\right]=2 \int_{t_{i}}^{t_{f}} d t \int_{t_{i}}^{t} d t^{\prime} \mathcal{R}\left[f(t) \bar{f}\left(t^{\prime}\right)\right]=\left|\int_{t_{i}}^{t_{f}} f(t) d t\right|^{2} \tag{5.18}
\end{equation*}
$$

The integral $O_{2}$ becomes:

$$
\begin{equation*}
O_{2}=-\frac{1}{2}\left[\int_{t_{i}}^{t_{f}} d t e(t)\right]^{2}-\int_{t_{i}}^{t_{f}} d t f(t) \int_{t_{i}}^{t} d t^{\prime} \bar{f}\left(t^{\prime}\right) \tag{5.19}
\end{equation*}
$$

### 5.3 Robust population transfer - Figure of merit

For the case of a population transfer to a target state $\left|\phi_{T}\right\rangle$ (of given angle $\theta$ and internal phase $\varphi$ ), the final phase is irrelevant, one can consider the figure of merit as

$$
\begin{align*}
J & =\left|\left\langle\phi_{T} \mid \phi_{\alpha, \delta}\left(t_{f}\right)\right\rangle\right|^{2} \\
& =\left(1+O_{1}+O_{2}+\cdots\right)\left(1+\bar{O}_{1}+\bar{O}_{2}+\cdots\right) \\
& =1+O_{2}+O_{1} \bar{O}_{1}+\bar{O}_{2}+\cdots \tag{5.20}
\end{align*}
$$

where it can be noticed that all the odd terms of error cancel. The deviation with respect to one of the second order of $J$ reduces to a single integral:

$$
\begin{equation*}
O_{2}+O_{1} \bar{O}_{1}+\bar{O}_{2}=-\left|\int_{t_{i}}^{t_{f}} f(t) d t\right|^{2} \tag{5.21}
\end{equation*}
$$

One notes the remarkable property that, when one considers the robustness with respect to solely $\alpha$ (i.e. $\delta=0$ ), then the integrals $O_{n}$ do not depend on the particular time-parametrization of $\theta(t)$ (in the case of a monotonic $\theta(t)$, assuming a function $\widetilde{\gamma}(\theta)$ ) since

$$
\begin{align*}
\int_{t_{i}}^{t} e(t) d t & =\frac{1}{2} \alpha \int_{t_{i}}^{t} d t \dot{\gamma} \sin ^{2} \theta=\frac{1}{2} \alpha \int_{\theta_{i}}^{\theta} d \theta \frac{d \widetilde{\gamma}}{d \theta} \sin ^{2} \theta,  \tag{5.22a}\\
\int_{t_{i}}^{t} f(t) d t & =\frac{1}{2} \alpha \int_{t_{i}}^{t} d t\left(\frac{1}{2} \dot{\gamma} \sin 2 \theta-i \dot{\theta}\right) e^{i \gamma}=\frac{1}{2} \alpha \int_{\theta_{i}}^{\theta} d \theta\left(\frac{1}{2} \frac{d \widetilde{\gamma}}{d \theta} \sin 2 \theta-i\right) e^{i \widetilde{\gamma}} . \tag{5.22b}
\end{align*}
$$

We have considered the situation of an increasing $\theta(t)$, i.e. $\dot{\theta}(t)>0$. The opposite situation (i.e. a decreasing $\theta(t), \dot{\theta}(t)<0$ ) would add a minus sign in the right hand sides of Eq. (5.22a) and of Eq. (5.22b).

One can thus design a trajectory $\widetilde{\gamma}(\theta)$ when $\delta=0$ [which also sets the pulse area according to (5.5)] omitting the time-dependence (or equivalently for any timedependence). We notice that the robustness with respect to $\delta$ (when $\alpha=0$ ) can be in principle next conducted, keeping the trajectory $\widetilde{\gamma}(\theta)$ previously derived, by exploiting the time-dependence of $\theta$, for instance expanding $\theta(t)$ as a Fourier series [39] (leading to a protocol on a finite time $t \in[0, T]$ ). We can also optimize the time dependence of $\theta$ by considering the minimization of the pulse energy or of the time of interaction (for a bounded pulse amplitude), see Subsection 5.5.2.

Alternatively, in the case of a monotonic $\gamma(t)$, assuming a function $\widetilde{\theta}(\gamma)$, when one considers the robustness with respect to solely $\alpha$ (i.e. $\delta=0$ ), then the integrals $O_{n}$ do not depend on the particular time-parametrization of $\gamma(t)$ since

$$
\begin{align*}
\int_{t_{i}}^{t} e(t) d t & =\frac{1}{2} \alpha \int_{t_{i}}^{t} d t \dot{\gamma} \sin ^{2} \theta=\frac{1}{2} \alpha \int_{\gamma_{i}}^{\gamma} d \gamma \sin ^{2} \widetilde{\theta},  \tag{5.23a}\\
\int_{t_{i}}^{t} f(t) d t & =\frac{1}{2} \alpha \int_{t_{i}}^{t} d t\left(\frac{1}{2} \dot{\gamma} \sin 2 \theta-i \dot{\theta}\right) e^{i \gamma}=\frac{1}{2} \alpha \int_{\gamma_{i}}^{\gamma} d \gamma\left(\frac{1}{2} \sin 2 \widetilde{\theta}-i \frac{d \widetilde{\theta}}{d \gamma}\right) e^{i \gamma} . \tag{5.23b}
\end{align*}
$$

We have considered the situation of an increasing $\gamma(t)$, i.e. $\dot{\gamma}(t)>0$. The opposite situation (i.e. a decreasing $\gamma(t), \dot{\gamma}(t)<0$ ) would add a minus sign in the right hand sides of Eq. (5.23a) and of Eq. (5.23b).

In a more general case, we have to consider two (continuous) functions

$$
\begin{cases}\tilde{\theta}_{+}(\gamma) & \text { for } \dot{\gamma} \geq 0, \gamma=\left[\gamma_{i}, \gamma_{m}\right]  \tag{5.24}\\ \tilde{\theta}_{-}(\gamma) & \text { for } \dot{\gamma}<0, \gamma=\left[\gamma_{f}, \gamma_{m}[ \right.\end{cases}
$$

with $\theta_{m}=\widetilde{\theta}_{+}\left(\gamma_{m}\right)=\widetilde{\theta}_{-}\left(\gamma_{m}\right)$ and the integral (5.23b) to be nullified become:

$$
\begin{align*}
0= & \frac{1}{2} \int_{\gamma_{i}}^{\gamma_{m}} d \gamma e^{i \gamma}\left(\sin 2 \widetilde{\theta}_{+}-2 \tilde{\theta}_{+}\right)-i\left(\theta_{m} e^{i \gamma_{m}}-\theta_{i} e^{i \gamma_{i}}\right) \\
& -\frac{1}{2} \int_{\gamma_{f}}^{\gamma_{m}} d \gamma e^{i \gamma}\left(\sin 2 \widetilde{\theta}_{-}-2 \widetilde{\theta}_{-}\right)+i\left(\theta_{m} e^{i \gamma_{m}}-\theta_{f} e^{i \gamma_{f}}\right) \\
= & \frac{1}{2} \int_{\gamma_{i}}^{\gamma_{m}} d \gamma e^{i \gamma}\left(\sin 2 \widetilde{\theta}_{+}-2 \widetilde{\theta}_{+}\right)-i\left(\theta_{f} e^{i \gamma_{f}}-\theta_{i} e^{i \gamma_{i}}\right) \\
& -\frac{1}{2} \int_{\gamma_{f}}^{\gamma_{m}} d \gamma e^{i \gamma}\left(\sin 2 \widetilde{\theta}_{-}-2 \widetilde{\theta}_{-}\right) \tag{5.25}
\end{align*}
$$

### 5.4 The unconstrained optimization problem

Let us consider the unconstrained (non-robust) optimization problem. In this case, optimizing with respect to the pulse area, to the energy, or to the duration leads to the same trajectory, as it is shown below.

Minimizing the pulse area

$$
\begin{equation*}
\mathcal{A}(\gamma, \theta)=\int_{t_{i}}^{t_{f}} d t \Omega(t)=\int_{t_{i}}^{t_{f}} d t \sqrt{\dot{\theta}^{2}+\dot{\gamma}^{2} \sin ^{2} \theta(t)} \tag{5.26}
\end{equation*}
$$

can be formulated by the Euler-Lagrange equations

$$
\begin{equation*}
\operatorname{grad} \mathcal{A}(\gamma, \theta)=0 \tag{5.27}
\end{equation*}
$$

where the gradient is defined as

$$
\operatorname{grad} \mathcal{A}(\gamma, \theta)=\left[\begin{array}{c}
\frac{\partial \mathcal{L}_{0}}{\partial \gamma}-\frac{d}{d t}\left(\frac{\partial \mathcal{L}_{0}}{\partial \dot{\gamma}}\right)  \tag{5.28a}\\
\frac{\partial \mathcal{L}_{0}}{\partial \theta}-\frac{d}{d t}\left(\frac{\partial \mathcal{L}_{0}}{\partial \dot{\theta}}\right)
\end{array}\right],
$$

with the Lagrangian $\mathcal{L}_{0}$ defined from the pulse area as the cost:

$$
\begin{equation*}
\mathcal{A}(\gamma, \theta)=\int_{t_{i}}^{t_{f}} d t \sqrt{\dot{\theta}^{2}+\dot{\gamma}^{2} \sin ^{2} \theta(t)} \equiv \int_{t_{i}}^{t_{f}} d t \mathcal{L}_{0}(\dot{\gamma}, \theta, \dot{\theta}) \tag{5.29}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
\frac{\partial \mathcal{L}_{0}}{\partial \theta}-\frac{d}{d t}\left(\frac{\partial \mathcal{L}_{0}}{\partial \dot{\theta}}\right)=0, \quad \frac{\partial \mathcal{L}_{0}}{\partial \gamma}-\frac{d}{d t}\left(\frac{\partial \mathcal{L}_{0}}{\partial \dot{\gamma}}\right)=0 \tag{5.30}
\end{equation*}
$$

The optimal solution, which satisfies the Euler-Lagrange equations, can be found by inspecting the integral (5.26): it is simply achieved for $\dot{\gamma}=0$ for any given $\dot{\theta}$ (note that $\dot{\theta}$ cannot be 0 to accomplish a given transfer), i.e. $\gamma=$ Const. This gives $\varphi=\pi / 2$ from (5.3c), $\dot{\theta}=\Omega$ from (5.3a), $\Delta=0$ from (5.3b), and $\mathcal{A}_{\min }=\int_{t_{i}}^{t_{f}} d t \Omega(t)=\int_{t_{i}}^{t_{f}} d t \dot{\theta}=$ $\int_{\theta_{i}}^{\theta_{f}} d \theta=\theta_{f}-\theta_{i}$ (assuming a monotonic $\theta(t)$ such that $\dot{\theta}>0$, i.e. $\left.\Omega(t)>0\right)$.

Minimizing the pulse energy

$$
\begin{equation*}
\mathcal{E}(\gamma, \theta)=\int_{t_{i}}^{t_{f}} d t \Omega^{2}(t)=\int_{t_{i}}^{t_{f}} d t\left(\dot{\theta}^{2}+\dot{\gamma}^{2} \sin ^{2} \theta\right) \tag{5.31}
\end{equation*}
$$

is for the same reason achieved when $\dot{\gamma}=0$, leading to the same trajectory $\gamma=$ Const. as for the minimization of the pulse area. The function $\theta(t)$ that minimizes the energy is given by the Euler-Lagrange equation

$$
\begin{equation*}
\operatorname{grad} \mathcal{E}(\theta)=0 \tag{5.32}
\end{equation*}
$$

with

$$
\begin{equation*}
\operatorname{grad} \mathcal{E}(\theta)=\frac{\partial \mathcal{L}_{0}}{\partial \theta}-\frac{d}{d t}\left(\frac{\partial \mathcal{L}_{0}}{\partial \dot{\theta}}\right) \tag{5.33}
\end{equation*}
$$

where the Lagrangian $\mathcal{L}_{0}$ defined from the pulse energy

$$
\begin{equation*}
\mathcal{E}(\theta)=\int_{t_{i}}^{t_{f}} d t \dot{\theta}^{2} \equiv \int_{t_{i}}^{t_{f}} d t \mathcal{L}_{0}(\dot{\theta}) \tag{5.34}
\end{equation*}
$$

This leads to

$$
\begin{equation*}
\ddot{\theta}=0 \tag{5.35}
\end{equation*}
$$

i.e. to a linear evolution of $\theta(t)$ :

$$
\begin{equation*}
\theta=\left(\theta_{f}-\theta_{i}\right) \frac{t-t_{i}}{t_{f}-t_{i}}+\theta_{i} \tag{5.36}
\end{equation*}
$$

which gives a constant pulse:

$$
\begin{equation*}
\Omega=\frac{\theta_{f}-\theta_{i}}{t_{f}-t_{i}} \tag{5.37}
\end{equation*}
$$

and the minimum energy:

$$
\begin{equation*}
\mathcal{E}_{\min }=\left(\frac{\theta_{f}-\theta_{i}}{t_{f}-t_{i}}\right)^{2}\left(t_{f}-t_{i}\right) \tag{5.38}
\end{equation*}
$$

For the time-minimization problem, we can reparametrize the trajectories by the arclength $d s=d t \Omega(t) / \Omega_{0}$, i.e. $s=s_{i}+\int_{t_{i}}^{t} d t^{\prime} \Omega\left(t^{\prime}\right) / \Omega_{0}$ and the TDSE becomes:

$$
i \hbar \frac{\partial}{\partial s}\left|\widetilde{\phi}_{0}(s)\right\rangle=\frac{\hbar}{2}\left[\begin{array}{cc}
-\widetilde{\Delta} & \Omega_{0}  \tag{5.39}\\
\Omega_{0} & \widetilde{\Delta}
\end{array}\right]\left|\widetilde{\phi}_{0}(s)\right\rangle, \quad \widetilde{\Delta}=\Delta \Omega_{0} / \Omega
$$

which is a control problem with a bounded control $\widetilde{\Omega}=\Omega_{0}$, for which minimizing the pulse area comes down to minimizing the duration $t_{f}-t_{i}$ for a given peak $\Omega_{0}$ of the control. It follows that, for the initial control system, the problem of minimizing the pulse area $\mathcal{A}$, is equivalent to minimizing the time under the constraint on the control $\Omega \leq \Omega_{0}$ (in fact the minimum time is achieved when the pulse reaches its bound at all times: $\left.\Omega=\Omega_{0}\right)$. The minimum time is thus $T_{\min }=\left(\theta_{f}-\theta_{i}\right) / \Omega_{0}$.

For instance for a complete population transfer from $t_{i}=0$ to $t_{f}=T$, requiring $\theta_{i}=0$ and $\theta_{f}=\pi$, we have: $\theta=\pi t / T, \Omega=\pi / T, \mathcal{A}_{\min }=\pi, \mathcal{E}_{\min }=\pi^{2} / T$ and $T_{\text {min }}=\pi / \Omega_{0}$.

### 5.5 Application of RIO for robustness with respect to field inhomogeneities $(\delta=0)$

### 5.5.1 Complete population transfer

Complete population transfer corresponds to $\theta_{i}=0$ and $\theta_{f}=\pi, \gamma_{i}=\varphi_{i}$. Equation (5.3c)

$$
\begin{equation*}
\cot \varphi=\dot{\widetilde{\gamma}} \sin \theta=\frac{\sin \tilde{\theta}}{\dot{\tilde{\theta}}} \tag{5.40}
\end{equation*}
$$

implies $\varphi_{i}=\varphi_{f}=\pi / 2$ for any $\dot{\widetilde{\gamma}}\left(\theta_{i}\right)$ and $\dot{\widetilde{\gamma}}\left(\theta_{f}\right)$. Nullification of the second order (5.21) reduces to the two (real) conditions for a trajectory $\widetilde{\gamma}(\theta)$ :

$$
\begin{equation*}
0=\int_{0}^{\pi} d \theta \cos \widetilde{\gamma} \sin ^{2} \theta=\int_{0}^{\pi} d \theta \sin \widetilde{\gamma} \sin ^{2} \theta \tag{5.41}
\end{equation*}
$$

or to the two conditions for a trajectory $\widetilde{\theta}(\gamma)$ :

$$
\begin{align*}
& \int_{\pi / 2}^{\gamma_{f}} d \gamma(\sin 2 \widetilde{\theta}-2 \widetilde{\theta}) \cos \gamma+2 \pi \sin \gamma_{f}=0  \tag{5.42a}\\
& \int_{\pi / 2}^{\gamma_{f}} d \gamma(\sin 2 \widetilde{\theta}-2 \widetilde{\theta}) \sin \gamma-2 \pi \cos \gamma_{f}=0 \tag{5.42b}
\end{align*}
$$

### 5.5.1.1 Determination of the optimal trajectory $\widetilde{\gamma}(\theta)$ for a monotonic $\theta(t)$

The problem can be formulated as an optimization problem: finding the trajectory $\widetilde{\gamma}(\theta)$ that minimizes the pulse area (5.5)

$$
\begin{equation*}
J(\widetilde{\gamma})=\int_{0}^{\pi} d \theta \sqrt{1+(\dot{\widetilde{\gamma}})^{2} \sin ^{2} \theta} \equiv \int_{0}^{\pi} d \theta \mathcal{L}_{0}(\widetilde{\gamma}, \theta) \tag{5.43}
\end{equation*}
$$

with $\dot{\widetilde{\gamma}} \equiv \frac{d \widetilde{\gamma}}{d \theta}$, under the two constraints (5.41) rewritten as

$$
\begin{align*}
& \psi_{1}(\widetilde{\gamma})=\int_{0}^{\pi} d \theta \cos \widetilde{\gamma} \sin ^{2} \theta \equiv \int_{0}^{\pi} d \theta \varphi_{1}(\widetilde{\gamma}, \theta)=0  \tag{5.44a}\\
& \psi_{2}(\widetilde{\gamma})=\int_{0}^{\pi} d \theta \sin \widetilde{\gamma} \sin ^{2} \theta \equiv \int_{0}^{\pi} d \theta \varphi_{2}(\widetilde{\gamma}, \theta)=0 \tag{5.44b}
\end{align*}
$$

Since the final phase is irrelevant for the population transfer problem, the final value $\gamma_{f} \equiv \widetilde{\gamma}\left(\theta_{f}\right)$ is not fixed.

The problem can be solved by the Lagrange multiplier method extended to the function space as follows: The trajectory $\widetilde{\gamma}$ is solution of

$$
\begin{equation*}
\operatorname{grad} J(\widetilde{\gamma})+\lambda_{1} \operatorname{grad} \psi_{1}(\widetilde{\gamma})+\lambda_{2} \operatorname{grad} \psi_{2}(\widetilde{\gamma})=0 \tag{5.45}
\end{equation*}
$$

with $\lambda_{j}, j=1,2$, the Lagrangian multipliers associated to the constraints, where the gradient is defined according to the Euler-Lagrange equation (which is zero without constraint):

$$
\begin{equation*}
\operatorname{grad} J(\widetilde{\gamma})=\frac{\partial \mathcal{L}_{0}}{\partial \widetilde{\gamma}}-\frac{d}{d \theta}\left(\frac{\partial \mathcal{L}_{0}}{\partial \dot{\tilde{\gamma}}}\right) \tag{5.46}
\end{equation*}
$$

This definition also applies for the constraints, $j=1,2$

$$
\begin{equation*}
\operatorname{grad} \psi_{j}(\widetilde{\gamma})=\frac{\partial \varphi_{j}}{\partial \widetilde{\gamma}}-\frac{d}{d \theta}\left(\frac{\partial \varphi_{j}}{\partial \dot{\tilde{\gamma}}}\right) \tag{5.47}
\end{equation*}
$$

We obtain:

$$
\begin{align*}
0 & =-\frac{d}{d \theta}\left(\frac{\partial \mathcal{L}_{0}}{\partial \dot{\tilde{\gamma}}}\right)+\lambda_{1} \frac{\partial \varphi_{1}}{\partial \widetilde{\gamma}}+\lambda_{2} \frac{\partial \varphi_{2}}{\partial \widetilde{\gamma}} \\
& =-\frac{d}{d \theta}\left(\frac{\dot{\tilde{\gamma}} \sin ^{2} \theta}{\sqrt{1+(\dot{\widetilde{\gamma}})^{2} \sin ^{2} \theta}}\right)+\sin ^{2} \theta\left(\lambda_{2} \cos \widetilde{\gamma}-\lambda_{1} \sin \widetilde{\gamma}\right) \\
& =\frac{\ddot{\tilde{\gamma}} \sin ^{2} \theta+\dot{\tilde{\gamma}} \sin 2 \theta}{\sqrt{1+(\dot{\tilde{\gamma}})^{2} \sin ^{2} \theta}}-\frac{(\dot{\widetilde{\gamma}})^{2} \sin ^{2} \theta\left(\ddot{\tilde{\gamma}} \sin ^{2} \theta+\frac{1}{2} \dot{\widetilde{\gamma}} \sin 2 \theta\right)}{\left[1+(\dot{\tilde{\gamma}})^{2} \sin ^{2} \theta\right]^{3 / 2}}-\sin ^{2} \theta\left(\lambda_{2} \cos \widetilde{\gamma}-\lambda_{1} \sin \widetilde{\gamma}\right), \tag{5.48}
\end{align*}
$$

which gives the equation for $\theta \neq 0$, after division by $\sin ^{2} \theta$ :

$$
\begin{align*}
0= & (\ddot{\tilde{\gamma}}+2 \dot{\widetilde{\gamma}} \operatorname{cotan} \theta)\left[1+(\dot{\widetilde{\gamma}})^{2} \sin ^{2} \theta\right]-(\dot{\tilde{\gamma}})^{2}\left(\ddot{\widetilde{\gamma}} \sin ^{2} \theta+\frac{1}{2} \dot{\widetilde{\gamma}} \sin 2 \theta\right) \\
& -\left(\lambda_{2} \cos \widetilde{\gamma}-\lambda_{1} \sin \widetilde{\gamma}\right)\left[1+(\dot{\widetilde{\gamma}})^{2} \sin ^{2} \theta\right]^{3 / 2} \tag{5.49}
\end{align*}
$$

which can be finally rewritten as

$$
\begin{equation*}
0=\ddot{\widetilde{\gamma}}+2 \dot{\widetilde{\gamma}} \operatorname{cotan} \theta+(\dot{\widetilde{\gamma}})^{3} \cos \theta \sin \theta-\left(\lambda_{2} \cos \widetilde{\gamma}-\lambda_{1} \sin \widetilde{\gamma}\right)\left[1+(\dot{\widetilde{\gamma}})^{2} \sin ^{2} \theta\right]^{3 / 2} \tag{5.50}
\end{equation*}
$$

or

$$
\begin{equation*}
0=\ddot{\widetilde{\gamma}}+\dot{\widetilde{\gamma}} \frac{\cos \theta}{\sin \theta}\left[2+(\dot{\widetilde{\gamma}})^{2} \sin ^{2} \theta\right]-\left(\lambda_{2} \cos \widetilde{\gamma}-\lambda_{1} \sin \widetilde{\gamma}\right)\left[1+(\dot{\widetilde{\gamma}})^{2} \sin ^{2} \theta\right]^{3 / 2} \tag{5.51}
\end{equation*}
$$

In order to lift the initial and final singularities of $\dot{\widetilde{\gamma}} \operatorname{cotan} \theta$, we multiplying this equation by $\sin \theta$ :

$$
\begin{equation*}
0=\ddot{\widetilde{\gamma}} \sin \theta+2 \dot{\widetilde{\gamma}} \cos \theta+(\dot{\widetilde{\gamma}})^{3} \cos \theta \sin ^{2} \theta-\sin \theta\left(\lambda_{2} \cos \widetilde{\gamma}-\lambda_{1} \sin \widetilde{\gamma}\right)\left[1+(\dot{\widetilde{\gamma}})^{2} \sin ^{2} \theta\right]^{3 / 2} \tag{5.52}
\end{equation*}
$$

and setting the limit $\theta \rightarrow 0$ (initial time) or $\theta \rightarrow \pi$ (final time) leads to $\dot{\widetilde{\gamma}}_{i}=0=\dot{\widetilde{\gamma}}_{f}$.

This corresponds to a system of first order ordinary differential equations:

$$
\begin{align*}
& \dot{\tilde{\gamma}}=y  \tag{5.53a}\\
& \dot{y}=\ddot{\widetilde{\gamma}}=-2 y \operatorname{cotan} \theta-y^{3} \cos \theta \sin \theta+\left(\lambda_{2} \cos \widetilde{\gamma}-\lambda_{1} \sin \widetilde{\gamma}\right)\left(1+y^{2} \sin ^{2} \theta\right)^{3 / 2} \tag{5.53b}
\end{align*}
$$

It is useful to consider the backward equation: $\hat{\gamma}(u)=\widetilde{\gamma}(\theta)$ with $u=\theta_{f}-\theta$, i.e. satisfying $\hat{\gamma}(0)=\widetilde{\gamma}\left(\theta_{f}\right)$ and $\hat{\gamma}\left(\theta_{f}\right)=\widetilde{\gamma}(0)$. This gives $\dot{\hat{\gamma}}=-\dot{\widetilde{\gamma}}$, and

$$
\begin{align*}
0= & \ddot{\hat{\gamma}}-2 \dot{\hat{\gamma}} \operatorname{cotan}\left(\theta_{f}-u\right)-(\dot{\hat{\gamma}})^{3} \cos \left(\theta_{f}-u\right) \sin \left(\theta_{f}-u\right) \\
& -\left(\lambda_{2} \cos \hat{\gamma}-\lambda_{1} \sin \hat{\gamma}\right)\left[1+(\dot{\hat{\gamma}})^{2} \sin ^{2}\left(\theta_{f}-u\right)\right]^{3 / 2} \tag{5.54}
\end{align*}
$$

This corresponds to a system of first order ordinary differential equations:

$$
\begin{align*}
& \dot{\hat{\gamma}}=y  \tag{5.55a}\\
& \dot{y}=\ddot{\hat{\gamma}}= 2 y \operatorname{cotan}\left(\theta_{f}-u\right)+y^{3} \cos \left(\theta_{f}-u\right) \sin \left(\theta_{f}-u\right) \\
&+\left(\lambda_{2} \cos \widetilde{\gamma}-\lambda_{1} \sin \widetilde{\gamma}\right)\left[1+y^{2} \sin ^{2}\left(\theta_{f}-u\right)\right]^{3 / 2} . \tag{5.55b}
\end{align*}
$$

We notice that when $\theta_{f}=\pi$ (complete population transfer), we obtain the differential
equation

$$
\begin{equation*}
0=\ddot{\hat{\gamma}}+2 \dot{\hat{\gamma}} \operatorname{cotan} u+(\dot{\hat{\gamma}})^{3} \cos u \sin u-\left(\lambda_{2} \cos \hat{\gamma}-\lambda_{1} \sin \hat{\gamma}\right)\left[1+(\dot{\hat{\gamma}})^{2} \sin ^{2} u\right]^{3 / 2} \tag{5.56}
\end{equation*}
$$

which is of the same form as the original one (5.50): this shows the symmetry of the trajectory around $\theta=\pi / 2$ for the problem of complete population transfer. By symmetry, we have thus

$$
\begin{equation*}
\dot{\tilde{\gamma}}(0)=\dot{\tilde{\gamma}}(\pi), \quad \ddot{\tilde{\gamma}}(0)=\ddot{\tilde{\gamma}}(\pi), \quad \widetilde{\gamma}(\pi / 2)=(\widetilde{\gamma}(\pi)+\widetilde{\gamma}(0)) / 2 . \tag{5.57}
\end{equation*}
$$

The symmetry implies that we can identify the differential equations at early and late times, i.e. in the limit $\theta \rightarrow 0$ :

$$
\begin{align*}
0 & =\ddot{\tilde{\gamma}}+2 \frac{\dot{\tilde{\gamma}}}{\sin \theta}-\left(\lambda_{2} \cos \widetilde{\gamma}_{i}-\lambda_{1} \sin \widetilde{\gamma}_{i}\right) \\
& =\ddot{\tilde{\gamma}}(\pi-\theta)+2 \frac{\dot{\tilde{\gamma}}(\pi-\theta)}{\sin (\pi-\theta)}-\left(\lambda_{2} \cos \widetilde{\gamma}_{f}-\lambda_{1} \sin \widetilde{\gamma}_{f}\right) \\
& =\ddot{\tilde{\gamma}}+2 \frac{\dot{\gamma}}{\sin \theta}-\left(\lambda_{2} \cos \widetilde{\gamma}_{f}-\lambda_{1} \sin \widetilde{\gamma}_{f}\right) \tag{5.58}
\end{align*}
$$

which, using $\widetilde{\gamma}_{i}=\pi / 2$, leads to

$$
\begin{equation*}
-\lambda_{1}=\lambda_{2} \cos \widetilde{\gamma}_{f}-\lambda_{1} \sin \widetilde{\gamma}_{f} \tag{5.59}
\end{equation*}
$$

We remark that the symmetry is not preserved for a partial transfer.

### 5.5.1.2 Determination of the optimal trajectory $\widetilde{\theta}(\gamma)$ for a monotonic $\gamma(t)$

The problem can be similarly formulated as an optimization problem: finding the trajectory $\widetilde{\theta}(\gamma)$ that minimizes the pulse area (5.6)

$$
\begin{equation*}
J(\widetilde{\theta})=\int_{\gamma_{i}}^{\gamma_{f}} d \gamma \sqrt{(\dot{\tilde{\theta}})^{2}+\sin ^{2} \widetilde{\theta}} \equiv \int_{\gamma_{i}}^{\gamma_{f}} d \gamma \mathcal{L}_{0}(\gamma, \widetilde{\theta}) \tag{5.60}
\end{equation*}
$$

with $\dot{\tilde{\theta}} \equiv \frac{d \widetilde{\theta}}{d \gamma}$, under the two constraints (5.42) rewritten for convenience as

$$
\begin{align*}
& \psi_{1}(\widetilde{\theta})=-\frac{1}{4} \int_{\gamma_{i}}^{\gamma_{f}} d \gamma(\sin 2 \widetilde{\theta}-2 \widetilde{\theta}) \sin \gamma \equiv \int_{\gamma_{i}}^{\gamma_{f}} d \gamma \varphi_{1}(\gamma, \widetilde{\theta})=-\frac{\pi}{2} \cos \gamma_{f},  \tag{5.61a}\\
& \psi_{2}(\widetilde{\theta})=\frac{1}{4} \int_{\gamma_{i}}^{\gamma_{f}} d \gamma(\sin 2 \widetilde{\theta}-2 \widetilde{\theta}) \cos \gamma \equiv \int_{\gamma_{i}}^{\gamma_{f}} d \gamma \varphi_{2}(\gamma, \widetilde{\theta})=-\frac{\pi}{2} \sin \gamma_{f} . \tag{5.61b}
\end{align*}
$$

Since the final phase is irrelevant for the population transfer problem, the final value $\gamma_{f}$ is not fixed, but $\theta_{f}=\widetilde{\theta}\left(\gamma_{f}\right)=\pi$. This can be solved by the Lagrange multiplier method: The trajectory $\widetilde{\theta}(\gamma)$ is solution of

$$
\begin{equation*}
\operatorname{grad} J(\widetilde{\theta})+\lambda_{1} \operatorname{grad} \psi_{1}(\widetilde{\theta})+\lambda_{2} \operatorname{grad} \psi_{2}(\widetilde{\theta})=0 \tag{5.62}
\end{equation*}
$$

with $\lambda_{j}, j=1,2$, the Lagrangian multipliers associated to the constraints, where

$$
\begin{equation*}
\operatorname{grad} J(\widetilde{\theta})=\frac{\partial \mathcal{L}_{0}}{\partial \widetilde{\theta}}-\frac{d}{d \gamma}\left(\frac{\partial \mathcal{L}_{0}}{\partial \dot{\tilde{\theta}}}\right), \tag{5.63}
\end{equation*}
$$

and $j=1,2$

$$
\begin{equation*}
\operatorname{grad} \psi_{j}(\widetilde{\theta})=\frac{\partial \varphi_{j}}{\partial \widetilde{\theta}}-\frac{d}{d \gamma}\left(\frac{\partial \varphi_{j}}{\partial \tilde{\tilde{\theta}}}\right) \tag{5.64}
\end{equation*}
$$

We obtain:

$$
\begin{align*}
0= & \frac{\partial \mathcal{L}_{0}}{\partial \widetilde{\theta}}-\frac{d}{d \gamma}\left(\frac{\partial \mathcal{L}_{0}}{\partial \dot{\tilde{\theta}}}\right)+\sum_{j=1,2} \lambda_{j}\left[\frac{\partial \varphi_{j}}{\partial \widetilde{\theta}}-\frac{d}{d \gamma}\left(\frac{\partial \varphi_{j}}{\partial \dot{\tilde{\theta}}}\right)\right] \\
= & \frac{\partial}{\partial \widetilde{\theta}} \sqrt{(\dot{\tilde{\theta}})^{2}+\sin ^{2} \widetilde{\theta}}-\frac{d}{d \gamma}\left(\frac{\partial}{\partial \dot{\tilde{\theta}}} \sqrt{(\dot{\tilde{\theta}})^{2}+\sin ^{2} \widetilde{\theta}}\right)-\frac{\lambda_{1}}{4} \frac{\partial}{\partial \widetilde{\theta}}[\sin \gamma(\sin 2 \widetilde{\theta}-2 \widetilde{\theta})] \\
& +\frac{\lambda_{2}}{4} \frac{\partial}{\partial \widetilde{\theta}}[\cos \gamma(\sin 2 \widetilde{\theta}-2 \widetilde{\theta})] \\
= & \frac{\sin \widetilde{\theta} \cos \tilde{\theta}-\ddot{\tilde{\theta}}}{\sqrt{(\dot{\tilde{\theta}})^{2}+\sin ^{2} \widetilde{\theta}}}+\frac{(\dot{\tilde{\theta}})^{2}(\ddot{\tilde{\theta}}+\sin \widetilde{\theta} \cos \widetilde{\theta})}{\left[(\dot{\tilde{\theta}})^{2}+\sin ^{2} \widetilde{\theta}\right]^{3 / 2}}-\frac{1}{2}(\cos 2 \widetilde{\theta}-1)\left(\lambda_{1} \sin \gamma-\lambda_{2} \cos \gamma\right), \tag{5.65}
\end{align*}
$$

i.e.

$$
\begin{align*}
0= & (\sin \tilde{\theta} \cos \tilde{\theta}-\ddot{\theta})\left[(\dot{\tilde{\theta}})^{2}+\sin ^{2} \widetilde{\theta}\right]+(\dot{\tilde{\theta}})^{2}(\ddot{\tilde{\theta}}+\sin \tilde{\theta} \cos \tilde{\theta}) \\
& +\sin ^{2} \widetilde{\theta}\left(\lambda_{1} \sin \gamma-\lambda_{2} \cos \gamma\right)\left[(\dot{\tilde{\theta}})^{2}+\sin ^{2} \widetilde{\theta}\right]^{3 / 2} \\
= & 2 \operatorname{cotan} \tilde{\theta}(\dot{\tilde{\theta}})^{2}+\sin \tilde{\theta} \cos \tilde{\theta}-\ddot{\tilde{\theta}}+\left(\lambda_{1} \sin \gamma-\lambda_{2} \cos \gamma\right)\left[(\dot{\tilde{\theta}})^{2}+\sin ^{2} \widetilde{\theta}\right]^{3 / 2}, \tag{5.66}
\end{align*}
$$

which finally gives the differential equation

$$
\begin{equation*}
\ddot{\tilde{\theta}}=2(\dot{\tilde{\theta}})^{2} \operatorname{cotan} \widetilde{\theta}+\sin \tilde{\theta} \cos \widetilde{\theta}+\left(\lambda_{1} \sin \gamma-\lambda_{2} \cos \gamma\right)\left[(\dot{\tilde{\theta}})^{2}+\sin ^{2} \widetilde{\theta}\right]^{3 / 2} \tag{5.67}
\end{equation*}
$$

It can be rewritten

$$
\begin{equation*}
\ddot{\tilde{\theta}}+\sin \tilde{\theta} \cos \tilde{\theta}=2\left((\dot{\tilde{\theta}})^{2}+\sin ^{2} \widetilde{\theta}\right)\left[\operatorname{cotan} \widetilde{\theta}+\left(\lambda_{1} \sin \gamma-\lambda_{2} \cos \gamma\right)\left((\dot{\tilde{\theta}})^{2}+\sin ^{2} \widetilde{\theta}\right)^{1 / 2}\right] \tag{5.68}
\end{equation*}
$$

The optimal robust trajectory $\tilde{\theta}_{\text {opt }}(\gamma)$, solution of (5.68), is obtained for the set of values of $\lambda_{1}$ and $\lambda_{2}$, which satisfies (5.61) (and we select the trajectory of smallest pulse area in case of more than one solution). We remark that the value of $\gamma_{f}$ results from this solution.

In order to analyze the initial and final singularities of $(\dot{\tilde{\theta}})^{2} \operatorname{cotan} \tilde{\theta}$, with $\dot{\tilde{\theta}}=1 / \dot{\tilde{\gamma}} \rightarrow$ $\infty$ and $\widetilde{\theta} \rightarrow 0$, $\pi$, we multiply this equation by $\sin \widetilde{\theta}$, and set $\widetilde{\theta} \rightarrow 0$ (initial time) with $\gamma=\pi / 2:$

$$
\begin{equation*}
\ddot{\tilde{\theta}} \sin \widetilde{\theta}=(\dot{\tilde{\theta}})^{2}\left(2+\lambda_{1} \dot{\tilde{\theta}} \sin \widetilde{\theta}\right) \tag{5.69}
\end{equation*}
$$

This shows that $\ddot{\tilde{\theta}}$ is initially infinite. This is identically the case finally.
This differential equation corresponds to a system of first order ordinary differential equations:

$$
\begin{align*}
& \dot{\tilde{\theta}}=y  \tag{5.70a}\\
& \dot{y}=\ddot{\tilde{\theta}}=2 y^{2} \operatorname{cotan} \widetilde{\theta}+\sin \widetilde{\theta} \cos \widetilde{\theta}+\left(\lambda_{1} \sin \gamma-\lambda_{2} \cos \gamma\right)\left(y^{2}+\sin ^{2} \widetilde{\theta}\right)^{3 / 2} \tag{5.70b}
\end{align*}
$$

We have formulated it such that one can recover this differential equation from the one (5.50), derived for the trajectory $\widetilde{\gamma}(\theta)$, by inverting the derivatives:

$$
\begin{align*}
& \dot{\tilde{\gamma}}=\frac{d \widetilde{\gamma}}{d \theta}=1 /\left(\frac{d \tilde{\theta}}{d \gamma}\right)=1 / \dot{\widetilde{\theta}}  \tag{5.71a}\\
& \frac{d}{d \theta}(\dot{\widetilde{\gamma}} \dot{\widetilde{\tilde{\theta}}})=0=\ddot{\widetilde{\gamma}} \dot{\tilde{\tilde{\theta}}}+\dot{\widetilde{\gamma}} \frac{d}{d \gamma}(\dot{\widetilde{\theta}}) \frac{d \widetilde{\gamma}}{d \theta}=\ddot{\tilde{\gamma}} \dot{\tilde{\theta}}+(\dot{\widetilde{\gamma}})^{2} \ddot{\tilde{\theta}} \tag{5.71b}
\end{align*}
$$

i.e.

$$
\begin{equation*}
\ddot{\tilde{\gamma}}=-\ddot{\tilde{\theta}} /(\dot{\tilde{\theta}})^{3} \tag{5.72}
\end{equation*}
$$

We indeed obtain from (5.50):

$$
\begin{equation*}
\ddot{\tilde{\theta}} /(\dot{\tilde{\theta}})^{3}=2 \operatorname{cotan} \tilde{\theta} / \dot{\tilde{\theta}}+\cos \tilde{\theta} \sin \tilde{\theta} /(\dot{\tilde{\theta}})^{3}-\left(\lambda_{2} \cos \gamma-\lambda_{1} \sin \gamma\right)\left[1+\sin ^{2} \widetilde{\theta} /(\dot{\tilde{\theta}})^{2}\right]^{3 / 2} \tag{5.73}
\end{equation*}
$$

which is (5.67) for $\theta \neq 0$.

The optimal robust trajectory is obtained by the numerical solution of (5.70) satisfying the constraints (5.61) for the values of $\lambda_{1} \approx-1.1150$ and $\lambda_{2} \approx-0.3047$ which lead to $\gamma_{f}=5 \pi / 3$ (this value is in fact approximately given with the method, the exact is given in [40] using Pontryagin's maximum principle in an extened Hilbert space). The obtained trajectory and the corresponding driving parameters, for a pulse resulting from the chosen parameterization

$$
\begin{equation*}
\gamma(t)=\frac{1}{2}\left(\gamma_{f}+\gamma_{i}\right)+\frac{1}{2}\left(\gamma_{f}-\gamma_{i}\right) \operatorname{erf}\left(\frac{t}{T}\right) \tag{5.74}
\end{equation*}
$$

are shown in Figure 5.2 and 5.3. We notice that the optimal robust trajectory $\widetilde{\theta}(\gamma)$ (see Figure 5.2) is a convenient representation since it forms a function contrary to the inverse one $\widetilde{\gamma}(\theta)$. We remark that we recover the robust optimal solution that has been derived in [40] by the PMP method in an extended Hilbert space.


Figure 5.2: Optimal robust trajectory $\widetilde{\theta}(\gamma)$ corresponding to $\lambda_{1} \approx-1.1150$ and $\lambda_{2} \approx$ -0.3047 (leading to $\gamma_{f}=5 \pi / 3$ ).


Figure 5.3: Time-dependence of Rabi frequency $\Omega$ (upper frame, solid blue line) and detuning $\Delta$ (upper frame, solid red line) and the resulting populations $P_{j}, j=1,2$ (lower frame) for complete population transfer for the time parameterization $\gamma(t)(5.74)$.

### 5.5.2 Robust trajectory optimal with respect to pulse energy or time

In this case, since the robustness integrals do not depend on the timeparametrization, optimizing with respect to the pulse area, to the energy, or to the duration leads to the same trajectory.

To prove it, let's assume we have determined the optimal trajectory $\widetilde{\gamma}(\theta)$ (possibly defined by multiple functions), solution of Eq. (5.50), that minimizes the pulse area (5.5) under the robustness constraints: nullification of Eq. (5.21). We now consider the energy minimization:

$$
\begin{equation*}
\mathcal{E}(\gamma, \theta)=\int_{t_{i}}^{t_{f}} d t\left(\dot{\theta}^{2}+\dot{\gamma}^{2} \sin ^{2} \theta\right) \equiv \int_{t_{i}}^{t_{f}} d t \mathcal{L}_{0}(\dot{\gamma}, \theta, \dot{\theta}) \tag{5.75}
\end{equation*}
$$

but rewritten in a time representation $\psi_{1}(\gamma, \theta)=\int_{t_{i}}^{t_{f}} \varphi_{1} d t$ and $\psi_{2}(\gamma, \theta)=\int_{t_{i}}^{t_{f}} \varphi_{2} d t$, with $\varphi_{1}=\dot{\theta} \cos \gamma \sin ^{2} \theta, \varphi_{2}=\dot{\theta} \sin \gamma \sin ^{2} \theta$, for consistency with the energy minimization representation. The trajectory $(\gamma(t), \theta(t))$ solution of the optimal problem can be formulated by the Euler-Lagrange equations:

$$
\begin{equation*}
\operatorname{grad} \mathcal{E}(\gamma, \theta)+\lambda_{1} \operatorname{grad} \psi_{1}(\gamma, \theta)+\lambda_{2} \operatorname{grad} \psi_{2}(\gamma, \theta)=0 \tag{5.76}
\end{equation*}
$$

with $\lambda_{j}, j=1,2$, the Lagrangian multipliers associated to the constraints, where the gradient is defined as

$$
\begin{align*}
& \operatorname{grad} \mathcal{E}(\gamma, \theta)=\left[\begin{array}{l}
\frac{\partial \mathcal{L}_{0}}{\partial \gamma}-\frac{d}{d t}\left(\frac{\partial \mathcal{L}_{0}}{\partial \dot{\gamma}}\right) \\
\frac{\partial \mathcal{L}_{0}}{\partial \theta}-\frac{d}{d t}\left(\frac{\partial \mathcal{L}_{0}}{\partial \dot{\theta}}\right)
\end{array}\right],  \tag{5.77a}\\
& \operatorname{grad} \psi_{j}(\gamma, \theta)=\left[\begin{array}{l}
\frac{\partial \varphi_{j}}{\partial \gamma}-\frac{d}{d t}\left(\frac{\partial \varphi_{j}}{\partial \dot{\gamma}}\right) \\
\frac{\partial \varphi_{j}}{\partial \theta}-\frac{d}{d t}\left(\frac{\partial \varphi_{j}}{\partial \dot{\theta}}\right)
\end{array}\right] . \tag{5.77b}
\end{align*}
$$

The Euler-Lagrange equations lead to

$$
\begin{align*}
0 & =\frac{\partial \mathcal{L}_{0}}{\partial \gamma}-\frac{d}{d t}\left(\frac{\partial \mathcal{L}_{0}}{\partial \dot{\gamma}}\right)+\sum_{j=1,2} \lambda_{j}\left[\frac{\partial \varphi_{j}}{\partial \gamma}-\frac{d}{d t}\left(\frac{\partial \varphi_{j}}{\partial \dot{\gamma}}\right)\right] \\
& =-2 \ddot{\gamma} \sin ^{2} \theta-2 \dot{\gamma} \dot{\theta} \sin 2 \theta-\dot{\theta} \sin ^{2} \theta\left(\lambda_{1} \sin \gamma-\lambda_{2} \cos \gamma\right) \tag{5.78a}
\end{align*}
$$

and

$$
\begin{align*}
0 & =\frac{\partial \mathcal{L}_{0}}{\partial \theta}-\frac{d}{d t}\left(\frac{\partial \mathcal{L}_{0}}{\partial \dot{\theta}}\right)+\sum_{j=1,2} \lambda_{j}\left[\frac{\partial \varphi_{j}}{\partial \theta}-\frac{d}{d t}\left(\frac{\partial \varphi_{j}}{\partial \dot{\theta}}\right)\right] \\
& =\dot{\gamma}^{2} \sin 2 \theta-2 \ddot{\theta}+\dot{\gamma} \sin ^{2} \theta\left(\lambda_{1} \sin \gamma-\lambda_{2} \cos \gamma\right) . \tag{5.78b}
\end{align*}
$$

Using

$$
\begin{equation*}
\ddot{\gamma}=\dot{\theta}^{2} \ddot{\tilde{\gamma}}+\ddot{\theta} \dot{\tilde{\gamma}}, \tag{5.79}
\end{equation*}
$$

Eq. (5.78a) reads

$$
\begin{equation*}
0=2\left(\ddot{\tilde{\gamma}}+\ddot{\theta} \ddot{\tilde{\gamma}} / \dot{\theta}^{2}\right)+4 \dot{\tilde{\gamma}} \operatorname{cotan} \theta+\frac{1}{\dot{\theta}}\left(\lambda_{1} \sin \gamma-\lambda_{2} \cos \gamma\right) \tag{5.80}
\end{equation*}
$$

which becomes, using (5.78b):

$$
\begin{equation*}
0=\ddot{\tilde{\gamma}}+2 \dot{\tilde{\gamma}} \operatorname{cotan} \theta+(\dot{\widetilde{\gamma}})^{3} \sin \theta \cos \theta+\frac{1}{2 \dot{\theta}}\left(\lambda_{1} \sin \gamma-\lambda_{2} \cos \gamma\right)\left[1+(\dot{\widetilde{\gamma}})^{2} \sin ^{2} \theta\right] . \tag{5.81}
\end{equation*}
$$

The latter is (5.50) if $\dot{\theta}$ satisfies:

$$
\begin{equation*}
\dot{\theta}=\frac{C}{\sqrt{1+(\dot{\widetilde{\gamma}})^{2} \sin ^{2} \theta}} \tag{5.82}
\end{equation*}
$$

with $C$ a constant. The $\lambda_{j}$ 's are indeed not identical between (5.50) and (5.81), but they produce both the same trajectory (since the constraints are the same) when (5.82) is satisfied. Equation (5.82) can be rewritten as

$$
\begin{equation*}
\int_{\theta_{i}}^{\theta} d \theta \sqrt{1+(\dot{\tilde{\gamma}})^{2} \sin ^{2} \theta}=C\left(t-t_{i}\right) \tag{5.83}
\end{equation*}
$$

or equivalently for a trajectory $\widetilde{\theta}(\gamma)$

$$
\begin{equation*}
\int_{\gamma_{i}}^{\gamma} d \gamma \sqrt{(\dot{\widetilde{\theta}})^{2}+\sin ^{2} \theta}=C\left(t-t_{i}\right) \tag{5.84}
\end{equation*}
$$

where the left-hand side is the partial pulse area (see Eq. (5.5)); the form of the righthand side implies that the pulse amplitude is in fact constant, i.e. $\Omega_{0} \equiv C$. We have thus shown that optimization with respect to the pulse area or to the pulse energy leads to the same trajectory $\widetilde{\gamma}(\theta)$ (or equivalently $\widetilde{\theta}(\gamma)$ ), and that the time-parametrization is such that the pulse amplitude is constant for the energy minimization.

The time-optimization for a bounded pulse amplitude $\Omega_{0}$ leads to the minimum time

$$
\begin{equation*}
T_{\min }=\frac{1}{\Omega_{0}} \int_{\gamma_{i}}^{\gamma_{f}} d \gamma \sqrt{(\dot{\tilde{\theta}})^{2}+\sin ^{2} \theta} \tag{5.85}
\end{equation*}
$$

For the complete population transfer designed in Subsection 5.5.1, the optimal time is determined as $T_{\min } \approx 5.84 / \Omega_{0}$. The corresponding detuning and populations for such robust time-optimal control for a flat pulse of $\Omega_{0}$ are shown in Figure 5.4. The detuning has the form of a complete period of the elliptic cosine

$$
\begin{equation*}
\Delta=\Delta_{0} \operatorname{cn}\left(\frac{4 K(m) t}{T_{\min }}+K(m), m\right), \quad t \in\left[0, T_{\min }\right], \tag{5.86}
\end{equation*}
$$

with $K(m)$ the complete elliptic integral of the first kind, $m=0.235$, and $\Delta_{0}=$ $8 K(m) \sqrt{m} / T_{\text {min }} \approx 1.114 \Omega_{0}$.


Figure 5.4: Resulting detuning and dynamics of the populations $P_{j}, j=1,2$, for robust time-optimal control [obtained for a flat pulse of Rabi frequency $\Omega_{0}$ according to (5.85)] showing the complete population transfer with the optimal time $T_{\min } \approx 5.84 / \Omega_{0}$.

### 5.5.3 Partial population transfer

One targets from the ground state $|0\rangle$ the half superposition state of internal (relative) phase $\varphi_{0}$ (up to an irrelevant global phase $\kappa$ )

$$
\begin{equation*}
\left|\phi\left(t_{f}\right)\right\rangle=\left|\phi_{T}\right\rangle \equiv \frac{1}{\sqrt{2}}\binom{1}{e^{-i \varphi_{0}}} e^{-i \kappa} \tag{5.87}
\end{equation*}
$$

which imposes the boundaries of $\theta$ as

$$
\begin{equation*}
\theta_{i}=0, \quad \theta_{f}=\pi / 2, \quad \gamma_{i}=\varphi_{i}, \quad \gamma_{f}=2 \kappa+\varphi_{0}, \quad \varphi_{f}=\varphi_{0} \tag{5.88}
\end{equation*}
$$

From Eq. (5.3c), we get as before $\varphi_{i}=\pi / 2$.

Nullification of the second order (5.21) reduces to the two (real) conditions for a trajectory $\widetilde{\gamma}(\theta)$ :

$$
\begin{equation*}
0=\int_{0}^{\pi / 2} d \theta \cos \widetilde{\gamma} \sin ^{2} \theta=\int_{0}^{\pi / 2} d \theta \sin \widetilde{\gamma} \sin ^{2} \theta \tag{5.89}
\end{equation*}
$$

or to the two conditions for a trajectory $\widetilde{\theta}(\gamma)$ :

$$
\begin{align*}
& \int_{\pi / 2}^{\gamma_{f}} d \gamma(\sin 2 \widetilde{\theta}-2 \widetilde{\theta}) \cos \gamma+\pi \sin \gamma_{f}=0  \tag{5.90a}\\
& \int_{\pi / 2}^{\gamma_{f}} d \gamma(\sin 2 \widetilde{\theta}-2 \widetilde{\theta}) \sin \gamma-\pi \cos \gamma_{f}=0 \tag{5.90b}
\end{align*}
$$

Similar analyses lead to the same expressions as Eqs. (5.53) and (5.70) for trajectories $\widetilde{\gamma}(\theta)$ and $\widetilde{\theta}(\gamma)$, respectively.

The obtained trajectory $\widetilde{\theta}_{\mathrm{opt}}(\gamma)$ (see Figure 5.5) corresponding to the values of $\lambda_{1} \approx-1.6974$ and $\lambda_{2} \approx-0.6465$ allows one to determine $\gamma_{f} \approx 1.48 \pi$ and $\dot{\widetilde{\gamma}}_{f}=0$, which, from Eq. (5.3c), gives the optimal relative phase $\varphi_{0}=\pi / 2$. The optimal robust trajectory and the resulting dynamics are shown in Figure 5.5 and 5.6, for

$$
\begin{equation*}
\gamma(t)=\frac{1}{2}\left(\gamma_{f}+\gamma_{i}\right)+\frac{1}{2}\left(\gamma_{f}-\gamma_{i}\right) \operatorname{erf}\left(\frac{t}{T}\right) \tag{5.91}
\end{equation*}
$$

We notice again that the optimal robust trajectory is a function $\widetilde{\theta}(\gamma)$ (see Figure 5.5) for a monotonic $\gamma(t)$, but that the inverse one $\widetilde{\gamma}(\theta)$ is not.

In the case of time optimization we obtain the optimal time $T_{\min } \approx 4.05 / \Omega_{0}$. Figure 5.7 demonstrates the resulting detuning and populations for robust time-optimal control. The detuning has the form of three-quarters of the elliptic cosine period

$$
\begin{equation*}
\Delta=\Delta_{0} \mathrm{cn}\left(\frac{3 K(m) t}{T_{\min }}+K(m), m\right), \quad t \in\left[0, T_{\min }\right] \tag{5.92}
\end{equation*}
$$

with $m=0.4$, and $\Delta_{0}=6 K(m) \sqrt{m} / T_{\min } \approx 1.66 \Omega_{0}$.


Figure 5.5: Optimal robust trajectory $\tilde{\theta}(\gamma)$ corresponding to $\lambda_{1} \approx-1.6974$ and $\lambda_{2} \approx$ -0.6465 (leading to $\gamma_{f} \approx 1.48 \pi$ ).


Figure 5.6: Time-dependence of Rabi frequency $\Omega$ (upper frame, solid blue line) and detuning $\Delta$ (upper frame, solid red line) and the resulting populations $P_{j}, j=1,2$ (lower frame) for partial population transfer targeting the half superposition state (up to a global phase).


Figure 5.7: Detuning and dynamics of the populations $P_{j}, j=1,2$, for robust timeoptimal control (for a flat pulse of Rabi frequency $\Omega_{0}$ ) showing the half superposition. We obtain the optimal time $T_{\min } \approx 4.05 / \Omega_{0}$.

### 5.6 Discussion

In this chapter, we have developed an inverse geometric optimization technique, referred to as RIO, which combines the Euler-Lagrange optimization and the inverseengineering SSSP method. They lead to the determination of trajectories which can be formulated with the constraints of robustness integrals and boundary conditions ensuring the exact transfer to a given target. The control fields are then derived from the obtained robust geodesics and the inverted dynamical equations.

We have shown the resulting exact-fidelity as two typical population profiles in Figures 5.3 and 5.6. The robustness, for instance, of targeting half superposition state, is depicted in Figure 5.8 (solid cyan lines). Comparing with the well known $\pi / 2$-pulse (see Figure 5.8, solid magenta lines), the transition probability shows, as expected, a flat profile with respect to small relative deviation $\alpha$ of the pulse amplitude (or area).


Figure 5.8: Comparison of the robustness profile given by the RIO trajectories (solid cyan lines) with $\pi / 2$-pulse (solid magenta lines). Upper frame: Transition probability as a function of $\alpha$. Lower frame: Logarithmic scale of the deviation targeting at half superposition state.

In addition, we emphasize that the technique of inverse optimization we have developed allows the design of the optimal and robust solutions of quantum control problems of the general form $H=H_{0}+\lambda V$, where robustness is meant with respect to $\lambda$. Its applicability necessitates the knowledge of the parametrization of the dynamics generated by $H_{0}$ and robustness is considered by perturbation of $\lambda V$. This includes low-dimensional dynamical symmetries for $H_{0}$, typically $\operatorname{SU}(2), \operatorname{SU}(3)$ [58] and $\operatorname{SU}(4)$ [59], for which dynamical invariants can be derived. But this does not limit the applicability to two-, three- or four-level systems; higher dimensions with specific symmetries can be considered [60]. the RIO method can treat robust optimization of the following multi-level problems:
(i) Stimulated Raman exact passage (STIREP) [61, 36] featuring a $\mathrm{SU}(2)$ symmetry in the resonant case, or more generally a $\operatorname{SU}(3)$ symmetry;
(ii) Two-qubit gate, represented as a four-level problem in its simplest form: Following Ref. [62], one can compensate the error in the phase of a two-qubit controlled-PHASE gate (e.g. implemented in an ion trap) using interactions of the form $T_{1} \equiv \frac{1}{2} \sigma_{x} \otimes \sigma_{x}$,
$T_{2} \equiv \frac{1}{2} \sigma_{x} \otimes \sigma_{y}$ and $T_{3} \equiv \frac{1}{2} \mathbb{1} \otimes \sigma_{z}$, which feature $\mathrm{SU}(2)$ symmetry;
(iii) Qudit gate (with an arbitrary dimension $d$ ), at the heart of quantum Fourier transform (a key ingredient of many quantum algorithms), in a multi-pod configuration with some overlapping controls [63] or with circulant symmetries [64].

The perturbation $\lambda V$ is not limited to imperfections of the driving pulse, but can also concern the leakage to undesirable states [65] or to a lossy environnement (for instance leading to dephasing noise [66]), where the latter problem takes the general form $L=L_{0}+\lambda V$ via Lindbladians. The application of the method can be then interpreted as a dynamical decoupling inverse optimization. Robustness and dynamical decoupling can also be treated simultaneously.

## Conclusion

In the first part of this thesis which constitutes Chapter 2, we introduce the invariantbased inverse engineering techniques, including basic relevant concepts (see Section 2.1), generalities and applications to two- and three-state systems (see Section 2.2), determination of exact passages in terms of single-mode and multi-mode driving (see Sections 2.3 and 2.4).

We show that the Lewis-Riesenfeld invariant allows the derivation of a mode that can be interpreted as an exact version of the ideal dark state leading to adiabatic passage (STIRAP). This mode induces an exact transfer instead of adiabatic and approximate and has to be considered in an inverse engineering procedure:

- taking the appropriate boundaries that ensure an exact transfer;
- choosing the dynamics of the angles.

This transfer is referred to as stimulated Raman exact passage (STIREP). We emphasize that, as expected, this mode that leads to an exact transfer features a non-zero transient component in the excited state contrary to the dark state. Minimizing such transient population in the excited state is of great significance since it will lead in practice to loss due to spontaneous emission.

With in mind that a method of low transient population in the excited state while achieving exact transfer is expected to feature a counterintuitive scheme (first Stokes and next pump pulses), we derive a simple sin / cos solution for the pump/Stokes pulses, respectively, using a simple linear parametrization of the angles in a multi-mode configuration.

In the second part (Chapters 3 and 4), we present the relevance and application of the techniques of optimal control theory (OCT) in quantum systems.

General introduction and main methods of OCT $[18,51]$ are presented in Sections 3.1-3.4, including the Mayer-Lagrange type as the general form of optimal control problems (see Section 3.1), the Lagrange multiplier method (see Section 3.2), the EulerLagrange principle (see Section 3.3) and the Pontryagin maximum principle (PMP) (see

Section 3.4). We show that the minimum time problems consist in optimizing bounded controls (see Section 3.5). Such problems and minimum pulse-energy problems can be treated using PMP and shooting methods (see Section 3.6). We observe that the solution derived from Section 3.6 (see Eq. (3.136)) is the opposite sequence with the same pulse shapes of the multi-mode driving given by Section 2.4 (see Eq. (2.49)). We also observe in this section that minimizing the pulse energy is equivalent to minimizing the transfer-time for bounded controls $u_{p}^{2}+u_{s}^{2} \leq$ const. We describe gradient method (see Section 3.7) and its application to $\Lambda$ systems (see Section 3.8). We indicate the properties and limits of the gradient method by the end of Section 3.7, such as leading to very different results depending on the algorithm and the initial condition used, rarely providing a global optimal solution, etc. Vivid depictions of such limits are shown in Figure 3.12 and Figure 3.13 .

In Chapter 4, we mainly apply PMP to investigate energy- (see Section 4.2) and time-minimum (see Section 4.3) optimizations under the constraint of a given admissible transient population in the upper state (via its time area) leading to exact state-to-state transfer of three-level $\Lambda$-type quantum systems. In the limit of relatively low loss of the upper state, we can identify such time area population in the upper state (in absence of loss) to the total loss of the system. Our study is thus referred to as optimal dissipative STIREP. We show the main numerical data in Table 4.2 and Table 4.4, and the following important results:

- when the given admissible transient population in the upper state is relatively large, we obtain an intuitive pulse sequence pump/Stokes (see, for instance, Figure 4.5 and Figure 4.20) as optimal controls, similar to the optimal controls without constraints (see Figure 4.2 and Figure 4.15);
- when the given admissible transient population in the upper state is low, we obtain as optimal controls (i) an overlapping pulse sequence of specific shapes (see Figures 4.7 - 4.8) for energy optimization and (ii) a shaping featuring a transient counterintuitive pulse between fast intuitive sequences for time optimization with constrained pulse amplitudes (see Figures 4.22-4.27).
This is unexpected since it is widely believed that adiabatic passage using an approximate dark state (via counterintuitive pulse sequence Stokes/pump) is the most efficient one for energy-minimal problems. This is not the case: application of PMP shows that it is more efficient to operate fast via the excited state than using long adiabatic passage accumulating lossy non-adiabatic population. The result shows that the pulses get shorter and more intense for lower admissible transient population in the upper state (corresponding to lower admissible loss in practice). On the other hand, for the minimum-time strategy, we show that the dynamics operates relatively close to a dark
state when the admissible loss is low.
We additionally compare the results of energy-optimal dissipative STIREP derived from Section 4.2 (see Eq. (4.101)) with standard STIRAP and parallel STIRAP (see Subsection 4.2.4), respectively, and observe its noticeable superiorities:
- in the situation of the same loss (i.e. the time area of the transient population in the excited state) with standard STIRAP, the pulse energy of energy-optimal dissipative STIREP is much smaller than standard STIRAP;
- in the situation of the same pulse energy with parallel STIRAP, the loss of optimal dissipative STIREP is significantly smaller than parallel STIRAP.
We also remark that, for relatively low loss which leads to two fully overlapping controls, energy-optimal dissipative STIREP can be transformed into a planar pendulum. The property that the optimal solution corresponds to an oscillating, rotating or separatrix trajectory depends on the value of the time area of the transient population in the excited state.

Making a comparison of these two types of optimal dissipative STIREP, we observe that (considering the same dissipation):

- for the energy-optimal dissipative STIREP, the pulse energy and the pulse area are smaller, and the peak pulse amplitude is larger, than for the time-optimal dissipative STIREP, for the same duration;
- for the energy-optimal dissipative STIREP, the pulse energy and pulse area are smaller, and the duration is longer, than for the time-optimal dissipative STIREP, for the same peak pulse amplitude;
- the controls for energy-optimal dissipative STIREP are less robust than timeoptimal dissipative STIREP considering a small systematic error on the control field (referred to as pulse inhomogeneities) when the given loss is low (see Figure 4.29 and Figure 4.30), but the controls derived from the time-optimal dissipative STIREP are not as robust as standard STIRAP, but more accurate (see Figure 4.30).

We next combine robustness with optimization in the last part of the thesis, Chapter 5. Instead of using a direct optimization procedure for OCT (as shown in Chapters 3 and 4), we develop a technique of geometric optimization that allows the derivation of optimal and robust solutions from an inverse optimization, named robust inverse optimization (RIO). We introduce the model with the dynamical variable space we study (see Section 5.1), the recipe and the application to determine complete and partial transfers that feature both optimality and robustness with respect to pulse inhomogeneities in terms of control pulse area (chosen as the cost) of RIO (see Sections 5.2-5.5).

The proposed method RIO combines the Euler-Lagrange optimization and the
inverse-engineering SSSP method with the following procedure:

- firstly applying the Euler-Lagrange optimization constrained by robustness integrals and by the boundaries ensuring exact, optimal and robust fidelity in the dynamical variable space;
- secondly deriving the control fields from the geodesic using the dynamical equation, i.e. the time-dependent Schrödinger equation for the present quantum control problem, formulated in an inverted way, in which we express the Hamiltonian elements (the controls) from the dynamical variables.

We conclude that RIO allows in principle the design of optimal and robust solutions of low-dimension quantum control problems. Resulting numerical trajectories for complete population transfers (see Figures $5.2-5.4$ ) and partial population transfers targeting half superpositions (see Figures 5.5-5.7) in two-state quantum systems have been shown.

Future works will apply RIO to other various quantum control problems such as quantum gates, other types of imperfections, Raman processes, or dynamical decoupling of undesirable effects. Furthermore, RIO will be applied to higher orders robustness, which is feasible with a convenient formulation of robustness integrals, defining additional dynamical variables when multiple integrals have to be considered.

## Bibliography

[1] J. P. Palao and R. Kosloff. Quantum computing by an optimal control algorithm for unitary transformations. Physical Review Letters, 89(18):188301, 2002. (Cited on page 1.)
[2] C. F. Roos, M. Chwalla, K. Kim, M. Riebe, and R. Blatt. 'Designer atoms' for quantum metrology. Nature, 443(7109):316-319, 2006. (Cited on page 1.)
[3] M. Pfender, P. Wang, H. Sumiya, S. Onoda, W. Yang, D. B. R. Dasari, P. Neumann, X. Y. Pan, J. Isoya, R. B. Liu, and J. Wrachtrup. High-resolution spectroscopy of single nuclear spins via sequential weak measurements. Nature Communications, 10(1):1-8, 2019. (Cited on page 1.)
[4] S. J. Glaser, T. Schulte-Herbrüggen, M. Sieveking, O. Schedletzky, N. C. Nielsen, O. W. Sørensen, and C. Griesinger. Unitary control in quantum ensembles: Maximizing signal intensity in coherent spectroscopy. Science, 280(5362):421-424, 1998. (Cited on page 1.)
[5] S. J. Glaser, U. Boscain, T. Calarco, C. P. Koch, W. Köckenberger, R. Kosloff, I. Kuprov, B. Luy, S. Schirmer, T. Schulte-Herbrüggen, D. Sugny, and F. K. Wilhelm. Training Schrödinger's cat: quantum optimal control. The European Physical Journal D, 69(12):279, 2015. (Cited on pages 1 and 2.)
[6] P. Doria, T. Calarco, and S. Montangero. Optimal control technique for manybody quantum dynamics. Physical Review Letters, 106(19):190501, 2011. (Cited on page 1.)
[7] D. Dong and I. R. Petersen. Quantum control theory and applications: a survey. IET Control Theory Applications, 4(12):2651-2671, 2010. (Cited on page 1.)
[8] C. Brif, R. Chakrabarti, and H. Rabitz. Control of quantum phenomena: past, present and future. New Journal of Physics, 12(7):075008, 2010. (Cited on page 1.)
[9] J. Oreg, F. T. Hioe, and J. H. Eberly. Adiabatic following in multilevel systems. Physical Review A, 29:690-697, 1984. (Cited on page 1.)
[10] C. E. Carroll and F. T. Hioe. Three-state systems driven by resonant optical pulses of different shapes. JOSA B, 5(6):1335-1340, 1988. (Cited on page 1.)
[11] N. V. Vitanov, A. A. Rangelov, B. W. Shore, and K. Bergmann. Stimulated Raman adiabatic passage in physics, chemistry, and beyond. Reviews of Modern Physics, 89:015006, 2017. (Cited on pages 1 and 2.)
[12] G. Dridi, S. Guérin, V. Hakobyan, H. R. Jauslin, and H. Eleuch. Ultrafast stimulated Raman parallel adiabatic passage by shaped pulses. Physical Review A, 80:043408, 2009. (Cited on pages 1, 2, 5 and 97.)
[13] S. Guérin and H. R. Jauslin. Control of quantum dynamics by laser pulses: Adiabatic floquet theory. Advances in Chemical Physics, 125:147-268, 2003. (Cited on pages 1 and 5.)
[14] D. Stefanatos and E. Paspalakis. Speeding up adiabatic passage with an optimal modified Roland-Cerf protocol. Journal of Physics A: Mathematical and Theoretical, 53(11):115304, 2020. (Cited on page 1.)
[15] K. Blekos, D. Stefanatos, and E. Paspalakis. Performance of superadiabatic stimulated Raman adiabatic passage in the presence of dissipation and OrnsteinUhlenbeck dephasing. Physical Review A, 102(2):023715, 2020. (Cited on page 1.)
[16] R. W. H. Sargent. Optimal control. Journal of Computational and Applied Mathematics, 124(1-2):361-371, 2000. (Cited on page 1.)
[17] L. S. Pontryagin. Mathematical Theory of Optimal Processes. Routledge, 2018. (Cited on pages 1, 31, 33 and 39.)
[18] D. D'Alessandro. Introduction to Quantum Control and Dynamics. CRC Press, 2007. (Cited on pages $1,22,28,31,32,33$ and 157.)
[19] N. Khaneja, R. Brockett, and S. J. Glaser. Time optimal control in spin systems. Physical Review A, 63(3):032308, 2001. (Cited on page 1.)
[20] U. Boscain, G. Charlot, J. P. Gauthier, S. Guérin, and H. R. Jauslin. Optimal control in laser-induced population transfer for two- and three-level quantum systems. Journal of Mathematical Physics, 43(5):2107-2132, 2002. (Cited on pages 1, 38, 39, 41 and 46.)
[21] B. Bonnard and D. Sugny. Optimal Control with Applications in Space and Quantum Dynamics. American Institute of Mathematical Sciences, 2012. (Cited on page 1.)
[22] S. A. Rice and M. Zhao. Optical Control of Molecular Dynamics. John Wiley, 2000. (Cited on page 1.)
[23] A. P. Peirce, M. A. Dahleh, and H. Rabitz. Optimal control of quantum-mechanical systems: Existence, numerical approximation, and applications. Physical Review A, 37:4950-4964, 1988. (Cited on page 1.)
[24] M. Dahleh, A. P. Peirce, and H. Rabitz. Optimal control of uncertain quantum systems. Physical Review A, 42:1065-1079, 1990. (Cited on page 1.)
[25] D. D'Alessandro and M. Dahleh. Optimal control of two-level quantum systems. IEEE Transactions on Automatic Control, 46(6):866-876, 2001. (Cited on page 1.)
[26] D. J. Egger and F. K. Wilhelm. Optimal control of a quantum measurement. Physical Review A, 90:052331, 2014. (Cited on page 1.)
[27] V. Martikyan, D. Guéry-Odelin, and D. Sugny. Comparison between optimal control and shortcut to adiabaticity protocols in a linear control system. Physical Review A, 101(1):013423, 2020. (Cited on page 1.)
[28] M. A. Nielsen and I. L. Chuang. Quantum Computation and Quantum Information. Cambridge University Press, 2010. (Cited on page 2.)
[29] S. Wimperis. Broadband, narrowband, and passband composite pulses for use in advanced NMR experiments. Journal of Magnetic Resonance, Series A, 109(2):221231, 1994. (Cited on page 2.)
[30] J. A. Jones. Designing short robust NOT gates for quantum computation. Physical Review $A, 87(5): 052317,2013$. (Cited on page 2.)
[31] G. T. Genov, D. Schraft, T. Halfmann, and N. V. Vitanov. Correction of arbitrary field errors in population inversion of quantum systems by universal composite pulses. Physical Review Letters, 113(4):043001, 2014. (Cited on page 2.)
[32] B. T. Torosov, S. Guérin, and N. V. Vitanov. High-fidelity adiabatic passage by composite sequences of chirped pulses. Physical Review Letters, 106(23):233001, 2011. (Cited on page 2.)
[33] D. Guéry-Odelin, A. Ruschhaupt, A. Kiely, E. Torrontegui, S. Martínez-Garaot, and J. G. Muga. Shortcuts to adiabaticity: Concepts, methods, and applications. Reviews of Modern Physics, 91(4):045001, 2019. (Cited on page 2.)
[34] E. Torrontegui, S. Ibánez, S. Martínez-Garaot, M. Modugno, A. del Campo, D. Guéry-Odelin, A. Ruschhaupt, X. Chen, and J. G. Muga. Shortcuts to adiabaticity. In Advances in Atomic, Molecular, and Optical Physics, volume 62, pages 117-169. Elsevier, 2013. (Cited on page 2.)
[35] D. Daems, A. Ruschhaupt, D. Sugny, and S. Guérin. Robust quantum control by a single-shot shaped pulse. Physical Review Letters, 111:050404, 2013. (Cited on pages 2, 14, 134 and 135.)
[36] X. Laforgue, X. Chen, and S. Guérin. Robust stimulated Raman exact passage using shaped pulses. Physical Review A, 100(2):023415, 2019. (Cited on pages 2, 14 and 154.)
[37] N. Khaneja, T. Reiss, C. Kehlet, T. Schulte-Herbrüggen, and S. J. Glaser. Optimal control of coupled spin dynamics: design of NMR pulse sequences by gradient ascent algorithms. Journal of Magnetic Resonance, 172(2):296-305, 2005. (Cited on pages 2 and 48.)
[38] T. Nöbauer, A. Angerer, B. Bartels, M. Trupke, S. Rotter, J. Schmiedmayer, F. Mintert, and J. Majer. Smooth optimal quantum control for robust solid-state spin magnetometry. Physical Review Letters, 115(19):190801, 2015. (Cited on page 2.)
[39] L. Van-Damme, D. Schraft, G. T. Genov, D. Sugny, T. Halfmann, and S. Guérin. Robust NOT gate by single-shot-shaped pulses: Demonstration of the efficiency of the pulses in rephasing atomic coherences. Physical Review A, 96(2):022309, 2017. (Cited on pages 2 and 137.)
[40] L. Van Damme, Q. Ansel, S. J. Glaser, and D. Sugny. Robust optimal control of two-level quantum systems. Physical Review A, 95:063403, 2017. (Cited on pages 2 and 146.)
[41] J. Zeng and E. Barnes. Fastest pulses that implement dynamically corrected singlequbit phase gates. Physical Review A, 98(1):012301, 2018. (Cited on page 2.)
[42] X. Chen, I. Lizuain, A. Ruschhaupt, D. Guéry-Odelin, and J. G. Muga. Shortcut to adiabatic passage in two- and three-level atoms. Physical Review Letters, 105:123003, 2010. (Cited on page 5.)
[43] X. Chen and J. G. Muga. Engineering of fast population transfer in three-level systems. Physical Review A, 86:033405, 2012. (Cited on page 5.)
[44] B. T. Torosov and N. V. Vitanov. Composite stimulated Raman adiabatic passage. Physical Review A, 87:043418, 2013. (Cited on page 5.)
[45] H. R. Lewis Jr and W. B. Riesenfeld. An exact quantum theory of the timedependent harmonic oscillator and of a charged particle in a time-dependent elec-
tromagnetic field. Journal of Mathematical Physics, 10(8):1458-1473, 1969. (Cited on page 6.)
[46] D. E. Kirk. Optimal Control Theory: An Introduction. Courier Corporation, 2004. (Cited on page 22.)
[47] F. H. Clarke. The generalized problem of Bolza. SIAM Journal on Control and Optimization, 14(4):682-699, 1976. (Cited on page 22.)
[48] C. Lanczos. The Variational Principles of Mechanics. University of Toronto Press, 1949. (Cited on page 24.)
[49] D. Sugny, A. Keller, O. Atabek, D. Daems, C. M. Dion, S. Guérin, and H. R. Jauslin. Reaching optimally oriented molecular states by laser kicks. Physical Review A, 69(3):033402, 2004. (Cited on page 25.)
[50] W. P. Healy. Non-Relativistic Quantum Electrodynamics. Academic Press, 1982. (Cited on page 29.)
[51] D. Sugny and C. Kontz. Optimal control of a three-level quantum system by laser fields plus von Neumann measurements. Physical Review A, 77(6):063420, 2008. (Cited on pages 36 and 157.)
[52] J. Barzilai and J. M. Borwein. Two-point step size gradient methods. IMA Journal of Numerical Analysis, 8(1):141-148, 1988. (Cited on page 51.)
[53] E. Assémat and D. Sugny. Connection between optimal control theory and adiabatic-passage techniques in quantum systems. Physical Review A, 86(2):023406, 2012. (Cited on page 70.)
[54] B. W. Shore. The Theory of Coherent Atomic Excitation, volume 1. New York: Wiley, 1990. (Cited on page 95.)
[55] B. Shore. Coherent manipulations of atoms using laser light. Acta Physica Slovaca. Reviews and Tutorials, 58(3):243-486, 2008. (Cited on page 95.)
[56] S. Guérin, S. Thomas, and H. R. Jauslin. Optimization of population transfer by adiabatic passage. Physical Review A, 65(2):023409, 2002. (Cited on page 95.)
[57] D. P. Bertsekas. Constrained Optimization and Lagrange Multiplier Methods. Athena Scientific, 1 edition, 1996. (Cited on page 131.)
[58] E. Torrontegui, S. Martínez-Garaot, and J. G. Muga. Hamiltonian engineering via invariants and dynamical algebra. Physical Review A, 89(4):043408, 2014. (Cited on page 154.)
[59] U. Güngördü, Y. Wan, M. A. Fasihi, and M. Nakahara. Dynamical invariants for quantum control of four-level systems. Physical Review A, 86(6):062312, 2012. (Cited on page 154.)
[60] F. T. Hioe. N-level quantum systems with $\mathrm{SU}(2)$ dynamic symmetry. JOSA B, $4(8): 1327-1332,1987$. (Cited on page 154.)
[61] V. Dorier, M. Gevorgyan, A. Ishkhanyan, C. Leroy, H. R. Jauslin, and S. Guérin. Nonlinear stimulated Raman exact passage by resonance-locked inverse engineering. Physical Review Letters, 119(24):243902, 2017. (Cited on page 154.)
[62] S. S. Ivanov and N. V. Vitanov. Composite two-qubit gates. Physical Review A, $92(2): 022333,2015$. (Cited on page 154.)
[63] B. Rousseaux, S. Guérin, and N. V. Vitanov. Arbitrary qudit gates by adiabatic passage. Physical Review A, $87(3): 032328$, 2013. (Cited on page 155.)
[64] P. A. Ivanov and N. V. Vitanov. Two-qubit quantum gate and entanglement protected by circulant symmetry. Scientific Reports, 10(1):1-11, 2020. (Cited on page 155.)
[65] F. Motzoi, J. M. Gambetta, P. Rebentrost, and F. K. Wilhelm. Simple pulses for elimination of leakage in weakly nonlinear qubits. Physical Review Letters, 103(11):110501, 2009. (Cited on page 155.)
[66] X. Lacour, S. Guérin, L. P. Yatsenko, N. V. Vitanov, and H. R. Jauslin. Uniform analytic description of dephasing effects in two-state transitions. Physical Review $A, 75(3): 033417,2007$. (Cited on page 155.)

Titre : Contrôle quantique optimal et robuste dans des systèmes de petite dimension
Mots clés : contrôle quantique, contrôle optimal, processus Raman stimulé

Résumé : La théorie du contrôle optimal (OCT) est une méthode pour obtenir les solutions optimales de systèmes quantiques contrôlés par des champs externes, fournissant un ensemble puissant d'outils et de concepts. L'un des objectifs de la thèse est d'adapter la technique OCT dans des systèmes quantiques à deux et trois états en tenant compte des pertes et de la robustesse, ce qui est primordial pour la mise en œuvre de techniques de contrôle dans une large classe de plateformes.
Sur la base de techniques d'ingénierie inverse et du principe du maximum de Pontryagin (PMP), nous établissons et testons les différentes stratégies optimales montrant comment contrôler le transfert dans des systèmes quantiques à trois niveaux en considérant des solutions optimales en énergie et en temps minimum en tenant compte des pertes. Ces résultats montrent en particulier que le passage
adiabatique habituel dans de tels systèmes, connu sous le nom de passage adiabatique Raman stimulé (STIRAP), qui conduit à un transfert imparfait, peut être rendu exact, réalisant ainsi le passage exact de Raman stimulé (STIREP) tout en réduisant l'énergie et la durée des contrôles.
Un des objectifs consiste à développer une nouvelle technique qui permet de combiner robustesse et optimisation. Plutôt que d'utiliser une procédure d'optimisation directe comme la technique OCT, nous développons une technique d'optimisation géométrique qui permet de dériver des solutions optimales et robustes à partir d'une optimisation inverse. La méthode appelée optimisation inverse robuste (RIO) permet d'obtenir des trajectoires numériques qui peuvent être rendues aussi précises que nécessaire. La méthode est polyvalente et peut être appliquée à divers types d'erreurs et de problèmes de contrôle quantique.

Title : Optimal and robust quantum control in low dimensional systems
Keywords : quantum control, optimal control, stimulated Raman processes


#### Abstract

Optimal control theory (OCT) is the basic and comprehensive method to obtain the optimal solutions of quantum systems controlled by external fields. It provides a powerful set of tools and concepts.One of the goals of the thesis is to design the technique of OCT in two- and three-state quantum systems taking into account losses and robustness, which is of primary importance for the implementation of control techniques in a broad class of platforms. Based on inverse-engineering techniques and the Pontryagin maximum principle (PMP), we establish and test the different optimal strategies showing how to control the transfer in threelevel quantum systems considering energy- and time-minimum optimal solutions taking into account losses. These results, in particular, show that the usual adiabatic passage in such systems,


known as stimulated Raman adiabatic passage (STIRAP), which leads to imperfect transfer, can be made exact thus achieving stimulated Raman exact passage (STIREP) while reducing the energy and the duration costs respectively of the controls.
We next combine robustness with optimization. Instead of using a direct optimization procedure from OCT, we develop a technique of geometric optimization that allows the derivation of optimal and robust solutions from an inverse optimization. The method named robust inverse optimization (RIO) allows one to obtain numerical trajectories that can be made as accurate as required. The method is versatile and can be applied to various types of errors and of quantum control problems.

