Optimal Control of Coupled Spin Dynamics under Cross-Correlated Relaxation

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Abstract

Relaxation effects impose fundamental limitations on our ability to coherently control quantum mechanical phenomena. In this paper, we solve a new class of optimal control problems, which helps establish physical limits on how closely a quantum mechanical system can be steered to a desired target state in the presence of relaxation. In particular, we explicitly compute the maximum coherence or polarization that can be transferred between coupled nuclear spins in the presence of very general decoherence mechanisms that include cross-correlated relaxation. We give analytical expressions for the optimal control laws (pulse sequences). Exploitation of cross-correlation effects has recently led to the development of powerful methods in NMR spectroscopy to study very large biomolecules in solution. We demonstrate that the optimal pulse sequences provide significant gains over these state of the art methods, opening new avenues for spectroscopy of much larger proteins. It is shown that in spite of very large relaxation rates, optimal control can transfer coherence without any loss when cross-correlated relaxation rates are tuned to auto-correlated relaxation rates.

1 Introduction

In this paper, we study some model control problems which arise in connection with optimal manipulation of dissipative quantum dynamics. It is shown that the questions about optimal control of quantum mechanical phenomenon in presence of dissipation are directly linked to optimization problems associated with a class of constrained bilinear control systems. These bilinear systems $\dot{x} = (A + \sum_i^n u_i B_i)x$ are characterized by the fact that the controls can be expressed as smooth functions of fewer parameters, i.e. $u_i = g_i(v_1, v_2, \ldots, v_k)$ where g_i are polynomials and $k < n$. A general study of these class of systems is expected to find immediate applications in coherent control of quantum mechanical phenomenon.

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In [5, 6], we studied in detail, the following constrained bilinear system. Consider the dynamical system

$$\frac{d}{dt}\begin{bmatrix} r_1 \\ r_2 \end{bmatrix} = \begin{bmatrix} -\alpha u_1^2 & -u_1 u_2 \\ u_1 u_2 & -\alpha u_2^2 \end{bmatrix} \begin{bmatrix} r_1 \\ r_2 \end{bmatrix};$$

Given $(r_1(0), r_2(0)) = (1, 0)$, find the optimal control $0 \leq u_1(t) \leq 1$, and $0 \leq u_2(t) \leq 1$ such that at the terminal time T, $r_2(T)$ is maximized. Problems of this nature arise naturally in the optimal control of quantum mechanical phenomenon in presence of relaxation. In this paper, we study a more general class of optimal control problems which arise in manipulation of coupled spin dynamics in NMR spectroscopy, in the presence of cross-correlated relaxation (as explained subsequently).

According to the postulates of quantum mechanics, the evolution of the state of a closed quantum system is unitary and is governed by Schrödinger equation. This evolution can be controlled by systematically changing the Hamiltonian of the system. The control of quantum systems has important applications in physics and chemistry. In particular, the ability to steer the state of a quantum system (or of an ensemble of quantum systems) from a given initial state to a desired target state forms the basis of fields of laser coherent control, quantum computing and spectroscopic techniques such as nuclear magnetic resonance (NMR) and electron spin resonance (ESR) spectroscopy. However, in all applications involving control and manipulation of quantum mechanical phenomenon, the system of interest is open, i.e. interacts with its environment (also termed lattice). This undesirable interaction with an external heat bath destroys phase correlations in the quantum system and relaxes the system to its equilibrium state. Manipulating quantum mechanical phenomenon in a manner that minimizes decoherence effects, is a practical problem, in the whole field of coherent spectroscopy and coherent control of quantum mechanical phenomenon.

In the field of NMR spectroscopy, sequence of radio-frequency (RF) pulses with well defined frequencies, amplitudes, phases, and durations are used to manipulate ensembles of spin systems. Applications range from NMR spectroscopy of biological macro molecules to the experimental implementation of quantum-computing algorithms. From a control perspective, the goal in these applications is to steer the

state of the spin system using the free-evolution Hamiltonian of the spin system (representing internal system dynamics) and using RF pulse sequences as control variables. Pulse-sequences should be designed to minimize the effects of relaxation or decoherence that are always present in practice. High relaxation or dissipation rates is a major bottleneck in NMR spectroscopy of large proteins. Therefore, developing methods for optimal control of quantum mechanical systems, which minimize dissipation effects is expected to have immediate impact on the field of coherent control of quantum mechanical systems.

The paper is organized as follows. In the following section we recapitulate the basics of quantum mechanics and dissipation in open quantum mechanical systems. In section 3, we look at the model problem of optimal control of coupled two spin system in presence of decoherence. We develop the main ideas presented in this paper through this example.

2 Control of Dissipative Quantum Dynamics

The state of a closed quantum system, represented by a vector $|\psi\rangle$, evolves unitarily, according to Schrödinger equation

$$\frac{d|\psi(t)\rangle}{dt} = -iH(t)|\psi(t)\rangle, \tag{1}$$

where $H(t)$ is the Hamiltonian of the system. In this paper, we will only be concerned with finite-dimensional quantum systems. We can split the Hamiltonian as $H = H_d + \sum_{j=1}^{m} u_j(t)H_j$, where H_d is the part of Hamiltonian that is internal to the system and we call it the drift or free evolution Hamiltonian and $\sum_{j=1}^{m} u_j(t)H_j$, is the part of the Hamiltonian that can be externally changed. H_j are the control or RF Hamiltonians and $u_j(t)$, the control fields. In this paper, we will focus on optimal control of ensembles of nuclear spins in NMR spectroscopy. The state of an ensemble of quantum mechanical systems is represented by its density matrix. Given an ensemble of quantum systems with the state vectors given by $|\psi_k\rangle$, $k = 1, \ldots, N$, respectively, the density matrix ρ is defined as

$$\rho = \frac{1}{N}\sum_{k=1}^{N} |\psi_k\rangle\langle\psi_k|,$$

where $\langle\psi_k|$ is the conjugate transpose of the vector $|\psi_k\rangle$. The density matrix of a closed quantum system then evolves as

$$\dot{\rho} = -i[H(t), \rho],$$

where $[,]$ is the matrix commutator. We will refer to the eigenvectors of H_d as the energy eigenstates.

For open quantum systems, the evolution of the system is no longer unitary. The density matrix of the system at any time t is related to the initial state $\rho(0)$ by a trace preserving map [2]

$$\rho(t) = \sum_k E_k(t)\rho(0)E_k^\dagger(t), \tag{2}$$

such that $\sum_k E_k(t)^\dagger E_k = I$ (The operators E_k are termed as Kraus operators). In general, it is not possible to write an evolution equation in time for the Kraus operators and hence the density matrix. However, in many practical applications of interest, the lattice can be approximated as an infinite thermostat, whose own state never changes. This assumption is also called Markovian approximation and under these assumptions, it is possible to write the evolution of the density matrix of the system alone in the form (Lindblad Form) [3]

$$\dot{\rho} = [-iH(t), \rho] + L(\rho), \tag{3}$$

where the term $L(\rho)$ models dissipation or relaxation. The relaxation term $L(\rho)$ is linear in ρ and has the general form

$$L(\rho) = \sum_\alpha J_\alpha[V_\alpha\rho V_\alpha^\dagger - \frac{1}{2}V_\alpha^\dagger V_\alpha\rho - \frac{1}{2}\rho V_\alpha^\dagger V_\alpha], \tag{4}$$

where the coefficients J_α contains the information about physical relaxation parameters (lifetimes, relaxation rates) and V_α denotes operators representing various relaxation mechanisms(Once a basis is chosen, V_α are just finite dimensional matrices).

Relaxation phenomenon is broadly classified into two categories. Adiabatic relaxation (decoherence) and non-adiabatic relaxation(dissipation). Let ρ_D be a density matrix which is diagonal in the energy eigenstates. All the super-operators V_β in equation (4) such that

$$[V_\beta\rho_D V_\beta^\dagger - \frac{1}{2}V_\beta^\dagger V_\beta\rho_D - \frac{1}{2}\rho_D V_\beta^\dagger V_\beta] = 0 \tag{5}$$

constitutes the adiabatic relaxation or the decoherence terms. This mode of relaxation doesn't effect the population of the energy eigenstates as no energy is exchanged between the environment and the system, but phase correlations between energy eigenstates is destroyed (represented by off diagonal terms in the density matrix when expressed in energy eigenstates). The super-operators V_β for which equation (5) is not satisfied, constitute non-adiabatic mode of relaxation in the system and brings the populations to equilibrium, while destroying off diagonal terms in the density matrix. Energy is exchanged between the system and the lattice.

Our focus, in this paper is on the relaxation phenomenon in the liquid state NMR spectroscopy. The case in which we are interested is well modeled as system consisting of two weakly interacting parts: the

spin system consisting of all spin degrees of freedom of the nuclei, and the lattice consisting of all other degrees of freedom of the liquid sample, associated with the molecular rotations and translations. Molecules in solution are constantly being bombarded with solvent molecules and undergo random "Brownian" motion as a result. This stochastic Brownian motion is the principle mechanism of relaxation in NMR spectroscopy. This small inter-collision time of the order of $10^{-14} - 10^{-12}$ seconds, ensures that the correlations between the spin system and the bath decay much faster than the evolution of the spin system and a Markovian approximation is a valid assumption.

To demonstrate the basic principles, we examine an isolated pair of heteronuclear spins I and S (spins that belong to different nuclear species) with indirect interaction (mediated by the surrounding electrons). For such a system, the Hamiltonian H_d is given by [1]

$$H_d = H_Z + H_{ind} \ , \tag{6}$$

where H_Z is the Zeeman Hamiltonian for the spins I and S.

$$H_Z = \omega_I I_z + \omega_S S_z \tag{7}$$

where $I_\alpha = \sigma_\alpha \otimes \mathbf{1}$ and $S_\beta = \mathbf{1} \otimes \sigma_\beta$, with $\alpha, \beta \in \{x, y, z\}$. The matrices $\sigma_x, \sigma_y, \sigma_z$ are the standard pauli matrices and $\mathbf{1}$ is the two dimensional identity matrix. H_{ind} is the Hamiltonian for the indirect interaction between the spins. The general form of H_{ind} for two spins is

$$H_{ind} = \sum_{\alpha,\beta} J_{\alpha,\beta} 2 I_\alpha S_\beta; \tag{8}$$

where $I_\alpha S_\beta = \sigma_\alpha \otimes \sigma_\beta$ and $\alpha, \beta \in \{x, y, z\}$. The only effective part of this interaction in liquids is its average over all relative orientations of the spins in space. In isotropic liquids it is of the form

$$H_{ind} = 2J\{I_x S_x + I_y S_y + I_z S_z\} \ , \tag{9}$$

where J is the scalar coupling constant. In the weak coupling limit ($J \ll |\omega_I - \omega_S|$), the indirect interaction Hamiltonian is simplified to the form [1]

$$H_{ind} = 2J I_z S_z \ . \tag{10}$$

For heteronuclear spins I and S the weak coupling condition is always satisfied. Thus, the deterministic Hamiltonian for our system is

$$H_0 = \omega_I I_z + \omega_S S_z + 2\pi J I_z S_z \ . \tag{11}$$

We know look at the various dissipation mechanisms for this system. In liquid solutions, the most important relaxation mechanisms are due to dipole-dipole interaction (DD) and chemical shift anisotropy (CSA), as well as their interference effects (e.g. DD-CSA cross correlation terms). Any magnetic nucleus in a molecule generates an instantaneous magnetic dipolar field that is proportional to the magnetic moment of the nucleus. As the molecule tumbles in solution, this field fluctuates and constitutes a mechanism for relaxation of nearby spins resulting from DD interaction. Chemical shifts are reflections of the electronic environment that modify the local magnetic fields experienced by different nuclei. These local fields are anisotropic; consequently, the components of the local fields vary as the molecule re orients as a result of molecular motion and lead to CSA relaxation mechanism. Cross-Correlated relaxation refers to interference effects between these two relaxation mechanisms. The Lindlad operator which captures these decoherence mechanisms takes the form,

$$\begin{aligned} L(\rho) \quad = \quad & k_{DD}[2I_zS_z,[2I_zS_z,\rho] + \ k^I_{CSA}[I_z,[I_z,\rho]] \\ & + \ k^S_{CSA}[S_z,[S_z,\rho]] + \ k^I_{DD/CSA}[2I_zS_z,[I_z,\rho]] \\ & + \ k^S_{DD/CSA}[2I_zS_z,[S_z,\rho]], \end{aligned} \tag{12}$$

The rates k_{DD}, k^I_{CSA}, k^S_{CSA} represent auto-correlated relaxation rates due to DD relaxation, CSA relaxation of spin I and CSA relaxation of spin S, respectively. The rates $k^I_{DD/CSA}$ and $k^S_{DD/CSA}$ represent cross-correlation rates of spin I and S caused by interference effects between DD and CSA relaxation. The relaxation rates depend on various physical parameters, such as the gyromagnetic ratios of the spins, the internuclear distance, the CSA tensors, the strength of the magnetic field and the correlation time of the molecular tumbling [1].

Let the initial density operator be $\rho(0) = A$ and denote the density operator at time t by $\rho(t)$. The maximum efficiency of transfer between A and target operator C is defined as the largest possible value of $\mathrm{trace}(C^\dagger \rho(t))$ for any time t (by convention operators A and C are normalized to norm 1).

In this paper, we address the problem of finding the maximum efficiency for the transfers

$$I_z \rightarrow 2I_zS_z \tag{13}$$

This transfer is of central importance for two-dimensional NMR spectroscopy. Let

$$k_a = k_{DD} + k^I_{CSA},$$

and

$$k_c = k^I_{DD/CSA},$$

denote the net auto-correlated and cross-correlated relaxation rates of spin I respectively.

We now make the following transformation (which corresponds to a doubly rotating frame). Let

$$U(t) = \exp(iH_Z t) = \exp[i(\omega_I I_z + \omega_S S_z)t], \tag{14}$$

and

$$\sigma(t) = U(t)\rho(t)U^{\dagger}(t). \tag{15}$$

then $\sigma(t)$ evolves as

$$\frac{d\sigma}{dt} = -iJ[2I_zS_z, \sigma] + L(\sigma)\ . \tag{16}$$

Now the problem is to steer $\sigma(0) = I_z$ in equation 16 as close to the final stae $2I_zS_z$.

3 Optimal control in the presence of cross-correlated relaxation

Remark 1 Let

$$x(t) = \begin{bmatrix} x_1(t) \\ x_2(t) \\ x_3(t) \\ x_4(t) \\ x_5(t) \\ x_6(t) \end{bmatrix} = \begin{bmatrix} \langle I_z\rangle(t) \\ \langle I_x\rangle(t) \\ \langle I_y\rangle(t) \\ \langle 2I_yS_z\rangle(t) \\ \langle 2I_xS_z\rangle(t) \\ \langle 2I_zS_z\rangle(t) \end{bmatrix},$$

where $\langle I_z(t)\rangle = trace(I_z\sigma(t))$ represents the expectation value of the operator I_z at time t.

Problem 1 Consider a control system

$$\dot{x}(t) = A(t)x(t),$$

where $A(t) =$

$$\begin{bmatrix} 0 & u(t) & v(t) & 0 & 0 & 0 \\ -u(t) & -k_a & 0 & -J & -kc & 0 \\ -v(t) & 0 & -k_a & -k_c & J & 0 \\ 0 & J & -k_c & -k_a & 0 & -v(t) \\ 0 & -k_c & -J & 0 & -k_a & -u(t) \\ 0 & 0 & 0 & v(t) & u(t) & 0 \end{bmatrix}. \tag{17}$$

Here $u(t)$ and $v(t)$ are the control terms corresponding to the rf field and k_a, k_c and J are positive constants. k_a represents the auto-correlation rate, k_c represents the cross-correlation rate and J represents strength of Hamiltonian coupling as described in previous section. Starting from $(x_1, x_2, x_3, x_4, x_5, x_6) = (1,0,0,0,0,0)$, what is the largest achievable value of x_6 and what is the optimal rf field $u(t)$ and $v(t)$ which achieves this transfer.

The main result of the paper is as follows.

Theorem 1 For the control system in problem 1, let

$$\zeta = \sqrt{\frac{k_a^2 - k_c^2}{k_c^2 + J^2}}.$$

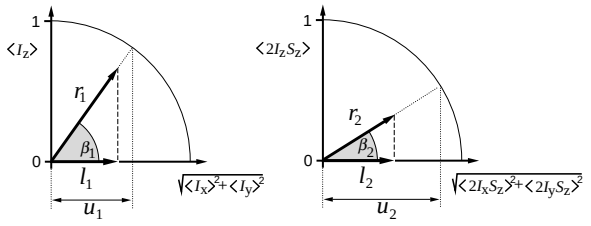

Figure 1: Representation of the system variables r_1, r_2, their transverse components l_1, l_2, the angles β_1, β_2, and of the control parameters $u_1 = \cos\beta_1$, $u_2 = \cos\beta_2$.

Starting from $(x_1, x_2, x_3, x_4, x_5, x_6) = (1,0,0,0,0,0)$, the largest achievable value of x_6 is

$$\eta = \sqrt{1+\zeta^2} - \zeta.$$

Let $\overline{\mathbf{R}((1,0,0,0,0,0))}$ denote the closure of the reachable set of the point $(1,0,0,0,0,0)$. All $(x_1, x_2, x_3, x_4, x_5, x_6)$ belonging to this set satisfy

$$\sqrt{\eta^2(x_1^2 + x_2^2 + x_3^2) + (x_4^2 + x_5^2 + x_6^2)} \leq \eta.$$

We develop the proof of this result in the remaining part of the paper.

Remark 2 We use $\vec{l_1}(t)$ and $\vec{l_2}(t)$ to denote the two dimensional vectors $(x_2(t), x_3(t))$ and $(x_5(t), x_4(t))$ respectively, as depicted in figures (1, 2). Let $r_1^2(t) = x_1^2(t) + x_2^2(t) + x_3^2(t)$ and $l_1^2(t) = x_2^2(t) + x_3^2(t)$. As shown in the figure (1), let $\cos\beta_1 = \frac{l_1(t)}{r_1(t)}$. Using rf fields (controls u and v in equation 17), we can control the angle β_1. Similarly let $l_2^2(t) = x_4^2(t) + x_5^2(t)$, and $r_2^2(t) = l_2^2 + x_6^2(t)$ with $\beta_2 = \cos^{-1}\frac{l_2}{r_2}$ (see Fig. 1). We can excercise control on the angle β_2 and we define $\cos\beta_2$ as a second control parameter u_2.

Remark 3 We superimpose the transverse planes defined by x_2 and x_3 with the plane defined by x_5 and x_4 such that x_2 is aligned with x_5 (Fig. 2). Let $\gamma(t)$ denote the angle between $\vec{l_1}(t)$ and $\vec{l_2}(t)$. Let

$$\theta = \tan^{-1}(\frac{J}{-k_c}).$$

In absence of control fields u and v, we have from Eq. (17),

$$\begin{aligned} \frac{d}{dt}\, l_1(t) &= -[\, k_a l_1(t) - \sqrt{k_c^2 + J^2}\cos(\theta+\gamma)\ l_2(t)] \\ \frac{d}{dt}\, l_2(t) &= -[\, k_a l_2(t) - \sqrt{k_c^2 + J^2}\cos(\theta-\gamma)\ l_1(t)]. \end{aligned}$$

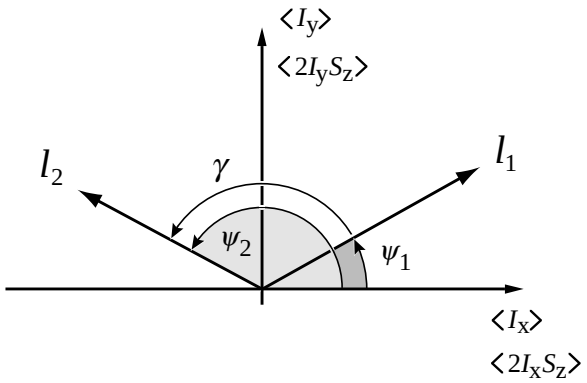

Figure 2: The figure shows the transverse planes defined by x_2 and x_3 superimposed on the plane defined by x_5 and x_4 such that x_2 is aligned with x_5 and x_4 aligned with x_3. Vectors $l_1(t)$ and $l_2(t)$ make an angle $\gamma(t)$ as shown in the figure.

This can be rewritten as $\frac{d}{dt}\begin{bmatrix} l_1(t) \\ l_2(t) \end{bmatrix} =$

$$J \begin{bmatrix} -\xi & \chi\ \cos(\theta+\gamma) \\ \chi\ \cos(\theta-\gamma) & -\xi \end{bmatrix} \begin{bmatrix} l_1(t) \\ l_2(t) \end{bmatrix}, \quad (18)$$

where

$$\xi = k_a/J$$

and

$$\chi = \sqrt{1+\left(\frac{k_c}{J}\right)^2}.$$

For a fixed value of β_1 and β_2, we get

$$\begin{aligned} \frac{d}{dt}r_1(t) &= \cos(\beta_1)\ \frac{d}{dt}l_1(t) \\ \frac{d}{dt}r_2(t) &= \cos(\beta_2)\ \frac{d}{dt}l_2(t). \end{aligned}$$

Substituting in 18, we then get $\frac{d}{dt}\begin{bmatrix} r_1(t) \\ r_2(t) \end{bmatrix} =$

$$J \begin{bmatrix} -\xi u_1^2 & \chi\ u_1u_2\cos(\theta+\gamma) \\ \chi\ u_1u_2\cos(\theta-\gamma) & -\xi u_2^2 \end{bmatrix} \begin{bmatrix} r_1(t) \\ r_2(t) \end{bmatrix}. \quad (19)$$

Remark 4 By use of controls u and v, we can change u_1, u_2 and γ, however we donot have independent control over these parameters. We for now relax this constrint and assume that u_1, u_2 and γ are independent controls. We will show that the optimal solution of this relaxed problem can also be achieved by appropriate choice of $u(t)$ and $v(t)$ in the original problem. We therefore first solve the following optimal control problem.

Problem 2 Given the dynamical system in equation (19), find the optimal control $u_1(t)$, $u_2(t)$, and $\gamma(t)$, so that starting from $(r_1(0), r_2(0)) = (1, 0)$, we achieve the largest value for r_2.

As remarked in the introduction, these are optimal control problems where the state enters linearly, whereas the controls can be expressed as smooth functions of fewer parameters.

Theorem 2 For the dynamical system in equation (19), the function

$$V(r_1, r_2) = \sqrt{\eta^2 r_1^2 + r_2^2},$$

is constant along the optimal trajectory and represents the optimal return function for the above problem with η as defined in theorem 1. The optimal trajectory has two invariants of motion. Along the optimal trajectory

$$\frac{l_2(t)}{l_1(t)} = \frac{u_2(t)r_2(t)}{u_1(t)r_1(t)} = \eta,$$

and the angle $\gamma(t)$ is maintained constant at

$$\gamma^* = \tan^{-1}\frac{1-\eta^2}{(1+\eta^2)\cot\theta}.$$

Proof: We use the maximum principle of pontryagin to compute the optimal control. The Hamiltonian $\mathbb{H}$ of the optimal control problem takes the form

$$\lambda_1(-\xi u_1^2 r_1 + \chi u_1 u_2 r_2 \cos(\theta+\gamma)) + \lambda_2(\chi\cos(\theta-\gamma)u_1u_2r_2 - \xi u_2^2 r_2),$$

where u_1, u_2, γ are the control variables and λ_1 and λ_2 are the costate variables. We introduce the following notation. Let

$$a = \frac{\lambda_2}{\lambda_1}; \quad b = \frac{r_2}{r_1}.$$

The Hamiltonian can then be written as

$$\mathbb{H} = -\lambda_1 r_1[\xi\, ab\, u_2^2 + \chi(b\cos(\theta+\gamma) + a\cos(\theta-\gamma))u_1u_2 + \xi u_1^2],$$

The pontryagin's maximum principle states that if u_1^*, u_2^*, γ^* are the optimal control laws then

$$\begin{aligned} (u_1^*, u_2^*, \gamma^*) &= \arg\max_{(u_1,u_2,\gamma)} \mathbb{H}(u_1, u_2, \gamma) \quad (20) \\ \mathbb{H}(u_1^*, u_2^*, \gamma^*) &= 0 \quad (21) \end{aligned}$$

If V is the optimal return function for the problem, then

$$(\lambda_1(t), \lambda_2(t)) = (\frac{\partial V}{\partial r_1}, \frac{\partial V}{\partial r_2})|_{(r_1^*(t), r_2^*(t))},$$

where $(r_1^*(t), r_2^*(t))$ is the optimal trajectory. Therefore for this problem, $\lambda_1(t) > 0$ and $\lambda_2(t) > 0$. Let

$$B = \chi(a\cos(\theta-\gamma) + b\cos(\theta+\gamma)).$$

Since $a, b > 0$, if $B \leq 0$ then the only solution to equations (20, 21) is the trivial solution $u_1^* = u_2^* = 0$. Therefore $B > 0$. Also note, when $B^2 < 4\xi^2 ab$, the

only solution to equations (20, 21) is again the trivial solution. Suppose $B^2 > 4\xi^2 ab$, i.e. $\frac{B^2}{4\xi^2} = ab + \kappa$ for $\kappa > 0$. Then

$$\mathbb{H} = -\lambda_1 r_1 [\frac{B}{2\sqrt{\xi}}\, u_2 - \sqrt{\xi} u_1]^2 + \kappa \lambda_1 r_1 u_1^2,$$

and hence $\max \mathbb{H} > 0$. Therefore the only case for which (20, 21) can be satisfied is

$$B^2 = 4ab\xi^2, \tag{22}$$

implying

$$\frac{1}{\eta}\cos(\theta - \gamma) + \eta \cos(\theta + \gamma) = 2\frac{\xi}{\chi}, \tag{23}$$

where $\eta = \sqrt{\frac{b}{a}}$. Maximizing $\mathbb{H}$ with respect to choice of γ gives that the optimal γ satisfies

$$\eta \sin(\theta + \gamma) - \frac{1}{\eta}\sin(\theta - \gamma) = 0, \tag{24}$$

Solving Eq (23, 24), then leads to

$$\eta = \sqrt{\zeta^2 + 1} - \zeta, \tag{25}$$

where $\zeta = \sqrt{\frac{k_a^2 - k_c^2}{J^2 + k_c^2}}$ and optimal $\gamma = \tan^{-1} \frac{1-\eta^2}{(1+\eta^2)\cot\theta}$.

Also maximizing $\mathbb{H}$, with respect to u_1 and u_2, we get

$$\frac{u_1^*}{u_2^*} = \frac{B}{2\xi}, \tag{26}$$

and from (22,23) that

$$\frac{u_2^* r_2}{u_1^* r_1} = \sqrt{1 + \zeta^2} - \zeta. \tag{27}$$

Using the optimal control law in eq (27) and (19), it is easily verified that $V(r_1, r_2) = \sqrt{r_2^2 + r_1^2 \eta^2}$ is constant along the system trajectories and is the optimal return function for the problem.

Remark 5 Observe, the optimal control law has two invariants of motion. Along the optimal trajectory

$$\frac{l_2(t)}{l_1(t)} = \eta,$$

and the angle $\gamma(t)$ is maintained constant at $\tan^{-1} \frac{1-\eta^2}{(1+\eta^2)\cot\theta}$. Consider the two dimentional vector $(v(t), u(t))$ in equation (17). Let $A^2(t) = u^2(t) + v^2(t)$, represent the amplitude of the rf field and let $\phi(t)$ denote the angle this vector makes relative to vector $\vec{l}_1$ ($\gamma - \phi$ relative to $\vec{l}_2$). We now determine expression for $A(t)$ and $\phi(t)$ from the invariants. Let $dl_1^\perp$ denote the change in vector $\vec{l}_1$ in direction perpendicular to the vector $\vec{l}_1$ by application of the rf-field in small time dt. Then observe

$$dl_1^\perp = Ax_1(t)\cos(\phi)dt.$$

Similarly

$$dl_2^\perp = Ax_6(t)\cos(\gamma - \phi)dt.$$

If $\frac{l_2}{l_1}$ is maintained at η then the angle γ does not change due to the evolution equation (18). Therefore we only need to consider the change in γ, due to the rf field. If γ is constant, then

$$\frac{dl_1^\perp}{l_1} = \frac{dl_2^\perp}{l_2}.$$

This gives $\tan(\beta_1)\cos(\phi) = \tan(\beta_2)\cos(\gamma^* - \phi)$ because $\frac{x_1}{l_1} = \tan\beta_1$ and $\frac{x_6}{l_2} = \tan\beta_2$. This then implies that

$$\phi = \tan^{-1}\left(\frac{\tan\beta_1}{\tan\beta_2 \sin\gamma^*} - \cot\gamma^*\right).$$

The amplitude can be determined from the condition that $\frac{l_2(t)}{l_1(t)}$ is maintained constant. This implies that $\frac{dl_1}{l_1} = \frac{dl_2}{l_2}$. Substituting

$$\frac{dl_1}{dt} = -\xi J l_1 + \chi J \cos(\theta + \gamma) l_2 + Ax_1 \sin(\phi)$$

and

$$\frac{dl_2}{dt} = -\xi J l_2 + \chi J \cos(\theta - \gamma) l_2 - Ax_6 \sin(\gamma - \phi),$$

we get

$$A = \frac{\left(\cos(\theta - \gamma^*) - \eta^2 \cos(\theta + \gamma^*)\right)\chi J}{\left(\tan\beta_1 \sin\phi + \tan\beta_2 \sin(\gamma^* - \phi)\right)\eta}.$$

The expressions of optimal A and ϕ are expressed in terms of the state of the system (angle β_1 and β_2) and we have a optimal feedback control law.

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