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Upper and lower bounds on the control field and the quality of achieved optimally controlled quantum molecular motion

Metin Demiralp [1] and Herschel Rabitz

Department of Chemistry, Princeton University, Princeton, NJ 08544-1009, USA

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A large class of problems in optimally controlled quantum or classical molecular dynamics has multiple solutions for the control field amplitude. A denumerably infinite number of solutions may exist depending on the structure of the design cost functional. This fact has been recently proved with the aid of perturbation theory by considering the electric field as the perturbating agent. In carrying out this analysis, an eigenvalue (i.e., a spectral parameter) appears which gives the degree of deviation of the control objective from its desired value. In this work, we develop a scheme to construct upper and lower bounds for the field amplitude and spectral parameter for each member of the denumerably infinite set of control solutions. The bounds can be tightened if desired. The analysis here is primarily restricted to the weak field regime, although the bounds for the strong field nonlinear case are also presented.

1. Introduction

In recent years, there has been considerable activity in the subject of understanding how optical electric fields may control or manipulate molecular scale events [1–24]. The meeting of desired molecular objectives with a control field is a matter of theoretical design or perhaps adaptive feedback in the laboratory. Various approaches have been suggested, with the most general being the introduction of systematic optimal design techniques [12–16]. In essence, the problem of design has an inverse nature, since the goal is to find the optical field in the Hamiltonian to meet the physical objective.

The inverse nature of control field design reveals that the problem is nonlinear, with the possibility of various distinct multiple control field solutions giving physically acceptable results. This matter has recently been explored within the framework of quantum and classically modelled dynamical systems [25–27]. Under rather mild assumptions, it was proved, in either case, that a denumerably infinite

[1] Permanent address: Istanbul Technical University, Faculty of Sciences and Letters, Engineering Sciences Department, Ayazağa Campus, Maslak, 80626 Istanbul, Turkey.

number of optimal control solutions may exist. Each solution may be thought of as yielding the desired objective to a measured degree, in balance with other penalties that enter. The latter quantities may typically include penalties against optical field fluence and the accessing of undesired molecular states. Various numerical illustrations of molecular control have verified the existence of multiple solutions, although little is known about the full family of solutions in any particular case. The present paper will further explore this topic, by developing upper and lower bounds for both the quality of the achieved molecular objective, as well as the control field amplitude, for an arbitrary number of the denumerably infinite set of solutions.

The approach taken in this work starts with quantum mechanical optimal control theory, and builds on earlier work establishing the multiplicity of solutions for these problems [25–27]. In particular, this latter work shows that the family of weak control fields could be obtained by solving a linear integral equation eigenvalue problem [28]. In doing so, the eigenvalues of this equation measure the deviation of the achieved control objective (i.e., the expectation value of the control operator) from its desired value. Starting with this formulation, upper and lower bounds for the so-called spectral parameters (eigenvalues) will be generated in the present work. These are bounds on the quality of the achieved control. In addition, the formulation will also be extended in a preliminary way to include bounds on the control field amplitudes, although additional complexity is involved in this situation. The present paper does not numerically explore the various bounds, but lays the groundwork for such studies.

The paper is organized as follows: Section 2 will present a summary of the relevant optimal design equations for whose solutions we seek upper and lower bounds. The bounds are developed based on the extensive use of specialized linear operator theory techniques, and for completion, the necessary background on this topic is presented in the appendices. Section 3 employs the techniques of the appendices to the molecular control problem, to provide explicit expressions for the bounds on the quality of the achieved control objective. Finally, section 4 presents some summarizing conclusions.

2. The field and the spectral equations

The goal of optimally controlling quantum dynamics phenomena reduces to the design problem of finding the control field $\mathcal{E}(t)$, such that the objectives are met in balance with various costs or penalties that may arise, subject to Schrödinger's equation being satisfied. This statement can be translated into a mathematical set of operations by defining a cost functional, to be minimized with respect to the unknown field, subject to Schrödinger's equation as a constraint. Details of this process can be found elsewhere, and especially in the prior work establishing the multiplicity of field solutions [25,26]; here we shall only summarize the key results

as a basis for developing the bounds later in the paper. The necessary Euler equations to be solved are summarized below.

$$i\hbar \frac{\partial \psi(t)}{\partial t} = [H_0 + \mu \mathcal{E}(t)]\psi(t), \quad \psi(0) = \tilde{\psi}, \tag{2.1a, b}$$

$$i\hbar \frac{\partial \lambda(t)}{\partial t} = [H_0 + \mu \mathcal{E}(t)]\lambda(t) - W_p(t)\langle \psi(t)|\hat{O}'|\psi(t)\rangle \hat{O}'\psi(t), \tag{2.2a}$$

$$\lambda(T) = \frac{i}{\hbar}\eta \hat{O}\psi(T), \tag{2.2b}$$

$$\mathcal{E}(t) = \frac{2}{W_{\mathcal{E}}(t)} \Re(\langle \lambda(t)|\mu|\psi(t)\rangle), \tag{2.3}$$

$$\langle \psi(T)|\hat{O}|\psi(T)\rangle = \tilde{O} + s\eta, \tag{2.4}$$

where $\psi(t)$ and $\lambda(t)$ are the wavefunction and the Lagrange multiplier function. The latter Lagrange multiplier function, satisfying eq. (2.2) enters to assure that the Schrödinger's equation in eq. (2.1) is properly included as a rigid constraint in the design problem. Here H_0 is the Hamiltonian of the free molecular motion, μ is the time-independent dipole function, and η is a parameter which measures the deviation of the objective term from its target value. The space-dependence of the entities ψ, $\tilde{\psi}$, λ, $\hat{O}$, $\hat{O}'$, and μ is implicitly understood. The operator $\hat{O}$ characterizes the objective whose expectation value is desired to approach a prescribed target value, $\tilde{O}$, and the expectation value of the operator $\hat{O}'$ is desired to be suppressed. The weight functions $W_p(t) > 0$ and $W_{\mathcal{E}}(t) > 0$ respectively are used in the penalty terms to suppress the expectation value of the undesired operator $\hat{O}'$ and to reduce the field amplitude. The symbol $\Re$ denotes the real part of a complex valued quantity, and s is a switching agent with values 0 or 1 according to the definition of the objective term. The flexible case $s = 1$ aims to optimally minimize the deviation in the expectation value of the objective operator, $\hat{O}$, from its target value $\tilde{O}$, while the constrained case where $s = 0$, has the goal of exact achievement to the target value. The latter case uses η as a Lagrange parameter to assure satisfaction of the target value constraint. By formally solving the optimal control equations above for $\psi(t)$ and $\lambda(t)$ the problem can be reduced to two coupled equations only containing $\mathcal{E}(t)$ and η as unknowns [26]. We call these cases "field" and "spectral" equations respectively as they determine the field $\mathcal{E}(t)$ and the spectral parameter η. The field equation has the form

$$\mathcal{M}\mathcal{E}(t) - \eta \mathcal{N}\mathcal{E}(t) = \eta r_1(t) + R_1(\mathcal{E}(t)) + \eta R_2(\mathcal{E}(t)), \tag{2.5}$$

where $r_1(t)$ is not dependent on the field amplitude and the spectral parameter, and the remainder terms $R_1(\mathcal{E}(t))$ and $R_2(\mathcal{E}(t))$ are purely nonlinear functionals in $\mathcal{E}$. The operators $\mathcal{M}$ and $\mathcal{N}$ are defined below:

$$\mathcal{M}f(t) \equiv \frac{1}{W_{\mathcal{E}}(t)} \int_0^t d\tau K_1(t,\tau) f(\tau) + \frac{1}{W_{\mathcal{E}}(t)} \int_t^T d\tau K_2(t,\tau) f(\tau) - f(t)\,, \tag{2.6}$$

$$\mathcal{N}f(t) \equiv \frac{1}{W_{\mathcal{E}}(t)} \int_0^t d\tau K_3(t,\tau) f(\tau) + \frac{1}{W_{\mathcal{E}}(t)} \int_t^T d\tau K_4(t,\tau) f(\tau)\,, \tag{2.7}$$

where $f(t)$ is assumed to lie in the space of square integrable functions over the time interval $[0, T]$ under the weight function $W_{\mathcal{E}}(t)$.

The kernels are given below:

$$K_j(t,\tau) = 2\Re(\kappa_j(\tau,t))\,, \quad j = 1,2,3,4\,, \tag{2.8}$$

where

$$\kappa_1(\tau,t) = v_1(\tau,t) + v_3(\tau,t) + v_5(\tau,t) + v_7(\tau,t)\,, \tag{2.9a}$$

$$\kappa_2(\tau,t) = v_2(\tau,t) + v_4(\tau,t) + v_6(\tau,t) + v_8(\tau,t)\,, \tag{2.9b}$$

$$\kappa_3(\tau,t) = v_9(\tau,t) + v_{11}(\tau,t)\,, \tag{2.9c}$$

$$\kappa_4(\tau,t) = v_{10}(\tau,t) + v_{11}(\tau,t)\,. \tag{2.9d}$$

The functions $v_i(\tau,t)$ are given in appendix A, and the function $r_1(t)$ has the form

$$r_1(t) = \frac{2}{W_{\mathcal{E}}(t)} \Im\{\langle\tilde{\psi}|Q(T)Q_\mu(t)|\tilde{\psi}\rangle\}\,, \tag{2.10a, b}$$

where $\Im$ denotes the imaginary part and Q, Q_μ are given in appendix A.

The linear integral operator appearing on the left hand side of the field equation (2.5) can be inverted through its eigenvalues and the eigenfunctions of the following problem:

$$\mathcal{M}e_k = \eta_k \mathcal{N}e_k\,, \quad k \geqslant 1\,; \quad (e_j, \mathcal{N}e_k) = \delta_{j,k}\,, \quad j,k \geqslant 1\,, \tag{2.11a, b}$$

where the scalar product with the functions $e_k(t)$ is defined as an integration over the interval $[0, T]$ under the weight $W_{\mathcal{E}}(t)$. Under rather general conditions the following relation can be written for the field:

$$\mathcal{E}(t) = \sum_{k=1}^{\infty} \frac{(e_k, r)}{\eta_k - \eta} e_k(t)\,, \quad r(t) = \eta r_1(t) + R_1(\mathcal{E}(t)) + \eta R_2(\mathcal{E}(t))\,. \tag{2.12a, b}$$

This relation is not an explicit solution for the field amplitude since its right side contains $\mathcal{E}(t)$. However $\mathcal{E}(t)$ only appears in the nonlinear contribution terms. Since these latter residual terms can be bounded, the spectral equation (2.12) is amenable to upper and lower bound construction on $\mathcal{E}(t)$ to be discussed in section 3.

The spectral equation can be written in the following general form:

$$(r_1, \mathcal{E}) + r_2 + R_3 + \eta R_4 = \tilde{O} + s\eta\,, \tag{2.13}$$

where R_3 and R_4 denote the remainder functionals which are purely nonlinear in $\mathcal{E}(t)$. The explicit structure of r_2 is

$$r_2 = \langle \tilde{\psi} | Q(T) | \tilde{\psi} \rangle . \tag{2.14}$$

If we substitute the field amplitude $\mathcal{E}(t)$ from eq. (2.12) into eq. (2.13), then we obtain the following form for the spectral equation:

$$\rho(\eta) - \left\{ s - R_4 + \frac{\tilde{O} - r_2 - R_3 - (\mathcal{E}_L, R_2)}{\eta} - \frac{(\mathcal{E}_L, R_1)}{\eta^2} \right\} = 0 , \tag{2.15}$$

where

$$\rho(\eta) = \sum_{k=1}^{\infty} \frac{(e_k, r_2)^2}{\eta_k - \eta} , \tag{2.16}$$

$$\mathcal{E}_L(t) = \sum_{k=1}^{\infty} \frac{(e_k, r_2) e_k}{\eta_k - \eta} . \tag{2.17}$$

Equations (2.12) and (2.15) are the corresponding coupled field and spectral equations. The spectral parameter η may be shown to generally take a denumerably infinite number of discrete values corresponding to there being an equivalent number of control fields $\mathcal{E}(t)$. Section 3 below will investigate placing bounds on the control field and spectral parameter, with an emphasis on the latter quantity.

3. Bounds for the spectral parameter, η, and the field amplitude, $\mathcal{E}(t)$

This section will draw on the material in section 2 and appendix B, to give bounds for the quality of achieved control, expressed in terms of the parameter η, as well as some comments on bounds for the control field amplitude. Emphasis will be placed on issues of the quality of achieved control.

3.1. OVERALL QUALITY OF CONTROL: A GLOBAL BOUND FOR THE LOCATION OF VERTICAL ASYMPTOTES η_k IN THE SPECTRAL EQUATION

The linear integral operators $\mathcal{M}$ and $\mathcal{N}$ are bounded if their kernels remain finite over the field-molecule interaction time ($t \in [0, T]$). Their kernels depend in a complex way on the physical inputs of the optimal control problem, however it is possible to show that they are Hilbert–Schmidt kernels or may be reconstructed to be of that form. Therefore we can assume that $\mathcal{M}$ and $\mathcal{N}$ have bounded kernels and $\mathcal{N}$ is positive (or negative) definite. The function $\rho(\eta)$ given by eq. (2.16) plays an important role for the determination of the spectral parameter values η. The multiplicity in η entirely comes from the existence of the vertical asymptotes η_k with

respect to η in the structure of $\rho(\eta)$. The location of these asymptotes encompassess a certain region of η values. To find a global bound for this overall region one can start with the eigenvalue equation given by (2.11a). Since $\mathcal{N}$ is assumed to be positive definite its square root can be defined. This enables us to write the following unit weighted eigenvalue equation:

$$\mathcal{N}^{-1/2}\mathcal{M}\mathcal{N}^{-1/2}\bar{e}_k = \eta_k \bar{e}_k\,, \quad k = 1, 2, \ldots\,. \tag{3.1}$$

The spectral theory of linear operators dictates that the spectrum of the above operator is bounded from above by its norm. Here we use the spectral norm which is defined as

$$\|\mathcal{N}^{-1/2}\mathcal{M}\mathcal{N}^{-1/2}\| \equiv \max_e \frac{(e, \mathcal{M}e)}{(e, \mathcal{N}e)}\,. \tag{3.3}$$

The operator $\mathcal{N}$ must be bounded from below since it is Hermitian and positive definite. If we denote its smallest eigenvalue by ν_0 then we have the following inequality:

$$(e, \mathcal{N}e) \geqslant \nu_0 (e, e)\,. \tag{3.4}$$

This immediately replaces eq. (3.3) with the following one:

$$\eta_H < \|\mathcal{N}^{-1/2}\mathcal{M}\mathcal{N}^{-1/2}\| \leqslant \frac{1}{\nu_0}\|\mathcal{M}\|\,, \tag{3.5a}$$

where η_H denotes the highest eigenvalue in eq. (3.1). On the other hand, a lower bound can be constructed by using the intervals given by eq. (B.7) and (B.8a, b) in appendix B as follows:

$$\eta_L > \min_e \{a_1 - \sqrt{a_2 - a_1^2}\}\,, \tag{3.5b}$$

$$a_1 = \frac{(e, \mathcal{N}^{-1/2}\mathcal{M}\mathcal{N}^{-1/2}e)}{(e, e)}\,, \tag{3.5c}$$

$$a_2 = \frac{(e, \mathcal{N}^{-1/2}\mathcal{M}\mathcal{N}^{-1}\mathcal{M}\mathcal{N}^{-1/2}e)}{(e, e)}\,. \tag{3.5d}$$

If the kernels of the operators $\mathcal{M}$ and $\mathcal{N}$ are slowly varying enough with time then the function $e(t)$ can be chosen as unity and the minimization in eq. (3.5b) becomes unnecessary. This is the generally expected behavior in the kernels of these operators as long as $\hat{O}$ and $\hat{O}'$ and dipole function are bounded. However, sufficient care must be paid to the structural analysis of these entities. Highly oscillatory behavior in the kernels may necessitate the use of different functions $e(t)$'s, (e.g., trigonometric functions instead of $e(t) = 1$). For this reason, a few linearly independent $e(t)$ functions should be used to assure the validity of the lower bound given above.

The upper bound in eq. (3.5a) can be evaluated through the determination of the lowest eigenvalue of $\mathcal{N}$. One can proceed to numerically evaluate the smallest eigenvalue of the operator $\mathcal{N}$ to any desired accuracy. This gives a numerical lower bound for ν_0 by setting the last meaningful nonzero digit of this approximate smallest eigenvalue to zero. Furthermore, the methods of appendix B can be applied to obtain a lower bound to ν_0. Therefore, a global bound for the region locating the vertical asymptotes of $\rho(\eta)$ can be constructed through rather simple numerical means.

3.2. QUALITY OF INDIVIDUAL CONTROL SOLUTIONS: BOUNDS FOR EACH SPECTRAL PARAMETER VALUE

The operator $\mathcal{N}^{-1/2}\mathcal{M}\mathcal{N}^{-1/2}$ is bounded as argued above. This means that its spectrum may have an accumulation point of eigenvalues at the origin of the complex plane. The possibility of the existence of an accumulation point at $\eta = 0$ implies that not only the linearized part of the optimal control problem under consideration, but its original nonlinear form may also have an accumulation point at $\eta = 0$. Therefore the exact achievement of the target value for the objective term may be possible depending on the structure of the cost functional of the problem under consideration.

Based on eq. (2.15) we define the following functions to facilitate the further analysis of the bounds:

$$\mathcal{S}_1(\eta) = s - R_4 + \frac{\tilde{O} - r_3 - R_3 - (\mathcal{E}_L, R_2)}{\eta} - \frac{(\mathcal{E}_L, R_1)}{\eta^2}\,, \tag{3.6a}$$

$$\mathcal{S}(\eta) = \rho(\eta) - \mathcal{S}_1(\eta)\,, \tag{3.6b}$$

where $\rho(\eta)$ and $\mathcal{E}_L$ are the functions given by eqs. (2.16) and (2.17). Since we have shown in appendix B how to construct bounds for the eigenvalues of a linear operator and the expansion coefficients of a given function with respect to this operator's eigenfunctions, we can assume that we *have upper and lower bounds* for the *eigenvalues*, η_k and the *residues*, (e_k, r_1). If we use the $\mathcal{B}_L$ and $\mathcal{B}_U$ functional symbols to denote these bounds we can write the following inequalities:

$$\mathcal{B}_L(\rho(\eta)) < \rho(\eta) < \mathcal{B}_U(\rho(\eta))\,, \tag{3.7}$$

$$\mathcal{B}_L(\mathcal{S}_1(\eta)) < \mathcal{S}_1(\eta) < \mathcal{B}_U(\mathcal{S}_1(\eta))\,, \tag{3.8}$$

where $\eta \in [\eta_k, \eta_{k+1}]$ and the explicit structure of the bounds are given below:

$$\mathcal{B}_L(\rho(\eta)) = \sum_{j=k+1}^{\infty} \frac{\mathcal{B}_L((e_j, r_1)^2)}{\mathcal{B}_U(\eta_j) - \eta} - \sum_{j=1}^{k} \frac{\mathcal{B}_U((e_j, r_1)^2)}{\eta - \mathcal{B}_U(\eta_j)}\,, \tag{3.9a}$$

$$\mathcal{B}_U(\rho(\eta)) = \sum_{j=k+1}^{\infty} \frac{\mathcal{B}_U((e_j, r_1)^2)}{\mathcal{B}_L(\eta_j) - \eta} - \sum_{j=1}^{k} \frac{\mathcal{B}_L((e_j, r_1)^2)}{\eta - \mathcal{B}_L(\eta_j)}, \tag{3.9b}$$

$$\mathcal{B}_L(\mathcal{S}_1(\eta)) = s - \mathcal{B}_U(R_4) + \frac{\tilde{O} - r_2 - \mathcal{B}_U(R_3) - \mathcal{B}_U((\mathcal{E}_L, R_2))}{\eta} - \frac{\mathcal{B}_U((\mathcal{E}_L, R_1))}{\eta^2}, \tag{3.10a}$$

$$\mathcal{B}_U(\mathcal{S}_1(\eta)) = s - \mathcal{B}_L(R_4) + \frac{\tilde{O} - r_2 - \mathcal{B}_L(R_3) - \mathcal{B}_L((\mathcal{E}_L, R_2))}{\eta} - \frac{\mathcal{B}_L((\mathcal{E}_L, R_1))}{\eta^2}. \tag{3.10b}$$

The bounds of the entities which are relevant to the nonlinear terms of the field and spectral equations are also functions of the spectral parameter, η. However, the bounded nature of the nonlinear terms allows for a relaxation of this problem. Henceforth we shall assume that $\mathcal{S}_1(\eta)$ is bounded from above and from below by η-independent entities, and $\mathcal{S}_{1,U}$, $\mathcal{S}_{1,L}$ will denote its upper and lower bounds respectively.

The upper and lower bounds given by eqs. (3.9a) and (3.9b) have meromorphic structures. In general, we will not be able to obtain analytic forms for the bounds, however the important aspect in the structure of these bounds is the number of poles. If we can reduce them to two poles then we can proceed to obtain analytical expressions for the solution of η in the interval, $[\eta_k, \eta_{k+1}]$ by using the standard formulae for the roots of a trinomial. Here, we shall give only the case of trinomial bounds.

If we assume again $\eta \in [\eta_k, \eta_{k+1}]$ then we can rewrite the analytic expression of $\rho(\eta)$ as follows:

$$\rho(\eta) = \sum_{j=k+1}^{\infty} \frac{(e_j, r_1)^2}{\eta_j - \eta} - \sum_{j=1}^{k} \frac{(e_j, r_1)^2}{\eta - \eta_j}. \tag{3.11}$$

This enables us to write the following inequality:

$$\left\{ \sum_{j=k+1}^{\infty} (e_j, r_1)^2 \right\} \frac{1}{\eta_{k+1} - \eta} - \frac{(e_k, r_1)^2}{\eta - \eta_k} > \rho(\eta) > \frac{(e_{k+1}, r_1)^2}{\eta_{k+1} - \eta} - \left\{ \sum_{j=1}^{k} (e_j, r_1)^2 \right\} \frac{1}{\eta - \eta_k}, \tag{3.12}$$

where we have only taken the two nearest poles to η. Since we have the following identity for the residues:

$$\sum_{j=k+1}^{\infty} (e_j, r_1)^2 = (r_1, \mathcal{N}^{-1} r_1) - \sum_{j=1}^{k} (e_j, r_1)^2 , \tag{3.13}$$

we can obtain the following two-pole formulae for the bounds of $\rho(\eta)$:

$$\frac{\rho_{up,r}}{\eta_{k+1} - \eta} - \frac{\rho_{up,l}}{\eta - \eta_k} > \rho(\eta) > \frac{\rho_{lw,r}}{\eta_{k+1} - \eta} - \frac{\rho_{lw,l}}{\eta - \eta_k} , \quad \eta \in [\eta_k, \eta_{k+1}] , \tag{3.14}$$

where

$$\rho_{up,r} = (r_1, \mathcal{N}^{-1} r_1) - \sum_{j=1}^{k} (e_j, r_1)^2 , \quad \rho_{up,l} = (e_k, r_1)^2 , \tag{3.15a, b}$$

$$\rho_{lw,r} = (e_{k+1}, r_1)^2 , \quad \rho_{lw,l} = \sum_{j=1}^{k} (e_j, r_1)^2 . \tag{3.15c, d}$$

Hence we can write

$$\mathcal{S}_U(\eta) > \mathcal{S}(\eta) > \mathcal{S}_L(\eta) , \tag{3.16}$$

where

$$\mathcal{S}_U(\eta) = \frac{\rho_{up,r}}{\eta_k - \eta} - \frac{\rho_{up,l}}{\eta - \eta_k} - \mathcal{S}_{1,L} , \quad \eta \in [\eta_k, \eta_{k+1}] , \tag{3.17a}$$

$$\mathcal{S}_L(\eta) = \frac{\rho_{lw,r}}{\eta_k - \eta} - \frac{\rho_{lw,l}}{\eta - \eta_k} - \mathcal{S}_{1,U} , \quad \eta \in [\eta_k, \eta_{k+1}] , \tag{3.17b}$$

Each of the functions $\mathcal{S}_U(\eta)$ and $\mathcal{S}_L(\eta)$ has only a single zero in the interval $[\eta_k, \eta_{k+1}]$. Thus, the solution of the spectral equation in this interval is bounded from above by the zero of $\mathcal{S}_L(\eta)$ and from below by the zero of $\mathcal{S}_U(\eta)$. Therefore we can write the following inequality:

$$\eta_{up} > \eta > \eta_{lw} , \tag{3.18}$$

where

$$\eta_{up} = \frac{\eta_{k+1} + \eta_k}{2} - \frac{\rho_{up,r} + \rho_{up,l}}{2\mathcal{S}_{1,L}} + \sqrt{\Delta_{up}} , \tag{3.19a}$$

$$\eta_{lw} = \frac{\eta_{k+1} + \eta_k}{2} - \frac{\rho_{lw,r} + \rho_{lw,l}}{2\mathcal{S}_{1,U}} + \sqrt{\Delta_{lw}} , \tag{3.19b}$$

and

$$\Delta_{up} = \left(\frac{\eta_{k+1} - \eta_k}{2} - \frac{\rho_{up,r} + \rho_{up,l}}{2\mathcal{S}_{1,L}} \right)^2 + \frac{\rho_{up,l}}{\mathcal{S}_{1,L}} (\eta_{k+1} - \eta_k) , \tag{3.20a}$$

$$\Delta_{lw} = \left(\frac{\eta_{k+1} - \eta_k}{2} - \frac{\rho_{lw,r} + \rho_{lw,l}}{2S_{1,U}} \right)^2 + \frac{\rho_{lw,l}}{S_{1,U}} (\eta_{k+1} - \eta_k) \,. \tag{3.20b}$$

A careful investigation shows that η_{lw} and η_{up} approache η_{k+1} when k tends to infinity. This means that the dominant terms in the asymptotic expansion of the solution of the spectral equation in the interval $[\eta_k, \eta_{k+1}]$ behave like η_{k+1} when k grows to infinity. Hence we can use η_{k+1} for the kth zero of the spectral equation as a first approximation.

The quantities $\rho_{up,l}$, $\rho_{up,r}$, $\rho_{lw,l}$, and $\rho_{lw,r}$ depend on the eigenfunctions, $e_k(t)$. To make the bounds more practical and get rid of this dependence we can relax these bounds either by discarding certain terms or by using the tools of appendix B for constructing the bounds to the eigenfunction expansion coefficients. This topic will not be pursued further.

3.3. BOUNDS FOR THE FIELD AMPLITUDE

We will now give a preliminary construction of the bounds for the field amplitude. As discussed in appendix B we will construct bounds not for the field amplitude itself but its inner product with a given function. This is sufficient since we can always express $\mathcal{E}(t)$ as an infinite sum of orthogonal functions as long as they span the space $\mathcal{E}(t)$ belongs to, except at the t-values where $\mathcal{E}(t)$ becomes singular. The construction of the set of these orthogonal functions is quite arbitrary except for the requirement of their completeness (and possibly compactness). One possible set is the $\{t^k\}_{k=0}^{\infty}$ basis set. The Gram–Schmidt orthonormalization procedure via use of an inner product under the weight function, $W_{\mathcal{E}}(t)$ enables us to obtain the desired orthonormalized basis functions.

Now we can write the following equalities for the inner product of the field amplitude with a given function, say $\gamma(t)$:

$$\begin{aligned} (\gamma, \mathcal{E}) = (\gamma, [\mathcal{M} - \eta\mathcal{N}]^{-1} r) &= (\gamma + r, [\mathcal{M} - \eta\mathcal{N}]^{-1} \{\gamma + r\}) \\ &\quad - (\gamma, [\mathcal{M} - \eta\mathcal{N}]^{-1} \gamma) - (r, [\mathcal{M} - \eta\mathcal{N}]^{-1} r) \,, \end{aligned} \tag{3.21}$$

where $r(t)$ was given by eq. (2.12b). The rightmost expression in these equalities involves the expectation value of the operator $[\mathcal{M} - \eta\mathcal{N}]^{-1}$ over the functions $\gamma(t) + r(t)$, $\gamma(t)$, and $r(t)$. The techniques [29–36] for the Stieltjes series and their Padé approximants can be used to construct bounds for these expectation values. However, we can also follow the same philosphy which was used to construct bounds for $\rho(\eta)$. The resultant meromorphic functions which have an infinite number of poles, may be replaced by simple structures having a finite number of poles. However, we can not avoid the use of the meromorphic functions whose poles are infinite or, at least, finite but higher in number, when we desire to tighten the lower and upper bounds. Another alternative for the construction of the bounds for $(\gamma, \mathcal{E})$ is the use of the approach given in appendix B.

4. Concluding remarks

In this work we aimed to construct bounds for the solutions of the field and the spectral equations. The spectral parameter, η, measures the deviation of the quantum control objective term from its target value. The main result in this case consists in demonstrating that explicit bounds can be constructed for η.

We have also proposed a scheme to construct the bounds for the inner product of the field amplitude with a known function rather than the field amplitude itself. The bounds for the solutions of the linearized field and spectral equations can be tightened without any complication. However, there is a limitation on the bound tightening procedure for the original nonlinear field and spectral equations because of the global structure of the nonlinear contributions to these equations. The methods introduced in this paper could be implemented numerically as a means for locating the η_k values and gaining insight into the multiplicity of control solutions.

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Appendix A

THE CONTROL KERNEL FUNCTIONS

The functions $v(\tau, t)$ entering eq. (2.9) are explicitly given below [25]:

$$v_1(\tau, t) = -\frac{1}{\hbar^2}\int_t^T d\tau_1 W_p(\tau_1)\langle\tilde{\psi}|Q_\mu(\tau)Q'(\tau_1)\tilde{P}Q'(\tau_1)Q_\mu(t)|\tilde{\psi}\rangle\,, \tag{A.1a}$$

$$v_2(\tau, t) = -\frac{1}{\hbar^2}\int_\tau^T d\tau_1 W_p(\tau_1)\langle\tilde{\psi}|Q_\mu(\tau)Q'(\tau_1)\tilde{P}Q'(\tau_1)Q_\mu(t)|\tilde{\psi}\rangle\,, \tag{A.1b}$$

$$v_3(\tau, t) = \frac{1}{\hbar^2}\int_t^T d\tau_1 W_p(\tau_1)\langle\tilde{\psi}|Q'(\tau_1)Q_\mu(\tau)\tilde{P}Q'(\tau_1)Q_\mu(t)|\tilde{\psi}\rangle\,, \tag{A.1c}$$

$$v_4(\tau, t) = \frac{1}{\hbar^2}\int_\tau^T d\tau_1 W_p(\tau_1)\langle\tilde{\psi}|Q'(\tau_1)Q_\mu(\tau)\tilde{P}Q'(\tau_1)Q_\mu(t)|\tilde{\psi}\rangle\,, \tag{A.1d}$$

$$v_5(\tau, t) = \frac{1}{\hbar^2}\int_t^T d\tau_1 W_p(\tau_1)\langle\tilde{\psi}|Q'(\tau_1)\tilde{P}Q_\mu(\tau)Q'(\tau_1)Q_\mu(t)|\tilde{\psi}\rangle\,, \tag{A.1e}$$

$$v_6(\tau,t)=\frac{1}{\hbar^2}\int_\tau^T d\tau_1\, W_p(\tau_1)\langle\tilde{\psi}|Q'(\tau_1)\tilde{P}Q_\mu(\tau)Q'(\tau_1)Q_\mu(t)|\tilde{\psi}\rangle\,, \tag{A.1f}$$

$$v_7(\tau,t)=\frac{1}{\hbar^2}\int_t^T d\tau_1\, W_p(\tau_1)\langle\tilde{\psi}|Q'(\tau_1)\tilde{P}Q'(\tau_1)Q_\mu(t)Q_\mu(\tau)|\tilde{\psi}\rangle\,, \tag{A.1g}$$

$$v_8(\tau,t)=\frac{1}{\hbar^2}\int_\tau^T d\tau_1\, W_p(\tau_1)\langle\tilde{\psi}|Q'(\tau_1)\tilde{P}Q'(\tau_1)Q_\mu(t)Q_\mu(\tau)|\tilde{\psi}\rangle\,, \tag{A.1h}$$

$$v_9(\tau,t)=\frac{1}{\hbar^2}\langle\tilde{\psi}|Q(T)Q_\mu(t)Q_\mu(\tau)|\tilde{\psi}\rangle\,, \tag{A.1i}$$

$$v_{10}(\tau,t)=\frac{1}{\hbar^2}\langle\tilde{\psi}|Q(T)Q_\mu(\tau)Q_\mu(t)|\tilde{\psi}\rangle\,, \tag{A.1j}$$

$$v_{11}(\tau,t)=\frac{1}{\hbar^2}\langle\tilde{\psi}|Q_\mu(\tau)Q(T)Q_\mu(t)|\tilde{\psi}\rangle\,, \tag{A.1k}$$

where

$$Q(t)=e^{\frac{i}{\hbar}tH_0}\hat{O}e^{-\frac{i}{\hbar}tH_0}\,,\quad Q(t)'=e^{\frac{i}{\hbar}tH_0}\hat{O}'e^{-\frac{i}{\hbar}tH_0}\,, \tag{A.2a, b}$$

$$Q_\mu(t)=e^{\frac{i}{\hbar}tH_0}\mu e^{-\frac{i}{\hbar}tH_0}\,,\quad \tilde{P}=|\tilde{\psi}\rangle\langle\tilde{\psi}|\,. \tag{A.2c, d}$$

Appendix B

SPECTRAL BOUNDS FOR LINEAR OPERATORS

This appendix presents some relevant aspects of spectral bounds for linear operators [29–31]. This is an encompassing subject in its own right, and only some relevant features utilized in section 3 will be summarized here. Consider a Hermitian, positive definite, bounded linear operator, $\mathcal{L}$, acting on a Hilbert space, $\mathcal{H}$. We assume that the spectrum of $\mathcal{L}$ is discrete, and the cartesian product of its eigenspaces is equivalent to $\mathcal{H}$. The eigenvalues and eigenfunctions of $\mathcal{L}$ are given below:

$$\mathcal{L}\psi_k=\lambda_k\psi_k\,,\quad 1\leqslant k\leqslant\infty\,, \tag{B.1}$$

where the eigenfunctions are ordered according to the eigenvalues $\lambda_1\leqslant\lambda_2\leqslant\lambda_3\ldots$ and the eigenfunctions are assumed to be orthonormal.

$$(\psi_j,\psi_k)=\delta_{jk}\,,\quad 1\leqslant j,k\leqslant\infty\,, \tag{B.2}$$

where δ_{jk} is the Kroenecker delta symbol.

B.1. Bounds for the eigenvalues

If we consider a real parameter, α, in the vicinity of the mth eigenvalue, λ_m, then the smallest eigenvalue of the operator $(\mathcal{L}-\alpha I)^2$ can be written as $(\lambda_m-\alpha)^2$. Hence the calculus of variations enables us to write the following relation for any function ϕ in $\mathcal{H}$:

$$(\phi,\mathcal{L}^2\phi)-2\alpha(\phi,\mathcal{L}\phi)+\alpha^2(\phi,\phi)\geqslant(\lambda_m-\alpha)^2(\phi,\phi)\,. \tag{B.3}$$

Although we assume that λ_m and ϕ are unknown, this inequality suffices for the evaluation of the upper and lower bounds for the mth eigenvalue of $\mathcal{L}$. We will present an approach to systematically determine α and ϕ to obtain bounds of any desired quality.

We can write the following bounds for λ_m based on eq. (B.3):

$$\alpha-\{(\langle\mathcal{L}\rangle-\alpha)^2+\langle\mathcal{L}^2\rangle-\langle\mathcal{L}\rangle^2\}^{1/2}\leqslant\lambda_m\leqslant\alpha+\{(\langle\mathcal{L}\rangle-\alpha)^2+\langle\mathcal{L}^2\rangle-\langle\mathcal{L}\rangle^2\}^{1/2}\,, \tag{B.4}$$

where $\langle\mathcal{L}\rangle$ and $\langle\mathcal{L}^2\rangle$ denote the expectation values of $\mathcal{L}$ and $\mathcal{L}^2$ with respect to ϕ,

$$\langle\mathcal{L}\rangle=\frac{(\phi,\mathcal{L}\phi)}{(\phi,\phi)}\,,\quad \langle\mathcal{L}^2\rangle=\frac{(\phi,\mathcal{L}^2\phi)}{(\phi,\phi)}\,. \tag{B.5a, b}$$

The fluctuation term, $\langle\mathcal{L}^2\rangle-\langle\mathcal{L}\rangle^2$ which is always positive, globally measures the deviation of ϕ from the eigenfunctions of $\mathcal{L}$. As ϕ approaches one of the eigenfunctions of $\mathcal{L}$, this term goes to zero. Hence, smaller values of the fluctuation imply tighter upper and lower bounds for the eigenvalues of $\mathcal{L}$. Therefore, we have the prospect of tightening the bounds in eq. (B.4) by inserting arbitrary parameters into the structure of ϕ and then minimizing the term $\langle\mathcal{L}^2\rangle-\langle\mathcal{L}\rangle^2$.

The real parameter α can be chosen as $\langle\mathcal{L}\rangle$ to minimize the bounds in (B.4):

$$\langle\mathcal{L}\rangle-\{\langle\mathcal{L}^2\rangle-\langle\mathcal{L}\rangle^2\}^{1/2}\leqslant\lambda_m\leqslant\langle\mathcal{L}\rangle-\{\langle\mathcal{L}^2\rangle-\langle\mathcal{L}\rangle^2\}^{1/2}\,. \tag{B.6a, b}$$

Now, the argument of the square root depends on ϕ and can be diminished by selecting an appropriate structure for ϕ. One rough but easy way to select ϕ is to replace it with the mth element of an orthonormal basis set which spans $\mathcal{H}$. If we denote this element by φ_m the relation above becomes

$$\langle\mathcal{L}\rangle_m-\{\langle\mathcal{L}^2\rangle_m-\langle\mathcal{L}\rangle_m^2\}^{1/2}\leqslant\lambda_m\leqslant\langle\mathcal{L}\rangle_m-\{\langle\mathcal{L}^2\rangle_m-\langle\mathcal{L}\rangle_m^2\}^{1/2}\,, \tag{B.7}$$

where

$$\langle\mathcal{L}\rangle_m=\frac{(\varphi_m,\mathcal{L}\varphi_m)}{(\varphi_m,\varphi_m)}\,,\quad \langle\mathcal{L}^2\rangle_m=\frac{(\varphi_m,\mathcal{L}^2\varphi_m)}{(\varphi_m,\varphi_m)}\,. \tag{B.8a, b}$$

These bounds define a denumerably infinite set of intervals for the eigenvalues of the operator $\mathcal{L}$. Although the desire for completely disjoint bounded intervals, they may overlap depending on the structure of the basis functions, φ_m. The suc-

cessful selection of these functions can be defined as the removal of the overlap among the intervals. For this task, one can use a linear combination of the φ functions instead of φ_m alone. For example, we can consider three neighboring φ-functions and employ a linear combination of three consecutive elements in the basis set $\varphi_{m-1}, \varphi_m, \varphi_{m+1}$ in place of φ_m alone. Then we can minimize the argument of the square root in (B.7) with respect to the coefficients of the linear combination. This procedure affects not only the argument of the square root term but $\langle \mathcal{L} \rangle_m$ at the same time. Therefore, both the measure and the location of the interval need consideration. The use of secondary or tertiary coupling effects in the construction of the linear combination for the basis function increases the number of the arbitrary coefficients to tighten the bounds.

The above procedure for optimally finding the basis functions will not be detailed here, but it can be developed rather straightforwardly. The actual choice of the basis functions is also an important matter, and better bounds will be obtained if knowledge of the operator $\mathcal{L}$ is built into the process. For example a Lanczos basis set may be formed by considering powers of the operator $\mathcal{L}$. Other means may also be employed, as appropriate [32–36].

B.2. Bounds for the coefficients in eigenfunction expansions

In spectral analysis, a bound on the expansion coefficients of a given function can be useful. If $\psi \in \mathcal{H}$ then we have the following eigenfunction expansion:

$$\psi = \sum_{j=1}^{\infty} (\psi_j, \psi)\psi_j \,. \tag{B.9}$$

To find bounds for (ψ_m, ψ), we can consider the following equality:

$$(\psi, [\mathcal{L} - \alpha\mathcal{I}]^{-2}\psi) = \frac{|(\psi_m, \psi)|^2}{(\lambda_m - \alpha)^2} + \sum_{\substack{j=1 \\ j \neq m}} \frac{|(\psi_j, \psi)|^2}{(\lambda_j - \alpha)^2} \,. \tag{B.10}$$

If we discard the infinite sum in this equation then we obtain the following upper bound for the mth coefficient of the expansion for ψ:

$$|(\psi_m, \psi)| < |\lambda_m - \alpha| (\psi, [\mathcal{L} - \alpha\mathcal{I}]^{-2}\psi)^{1/2} \,. \tag{B.11}$$

The lower bound for the same entity is a bit more complicated. It can be obtained by reducing the denominator values to a common smallest value in the infinite sum of eq. (B.10) and by using an identity over the resultant series of projection operators which creates the image of their object in the corresponding eigenspace of $\mathcal{L}$. The result is

$$|(\psi_m, \psi)| > \{\alpha_1 (\psi, [\mathcal{L} - \alpha\mathcal{I}]^{-2}\psi) - \alpha_2 (\psi, \psi)\}^{1/2} \,, \tag{B.12}$$

where

$$\alpha_1 = (\lambda_m - \alpha)^2 [\min_{j=m-1,m+1} \{(\lambda_j - \alpha)^2\} - (\lambda_m - \alpha)^2]^{-1} ,$$

$$\alpha_2 = \frac{\alpha_1}{\min_{j=m-1,m+1}\{(\lambda_j - \alpha)^2\}} . \qquad \text{(B.13a, b)}$$

Equations (B.11) and (B.12) are the desired upper and lower bounds for the eigen-expansion coefficients of a given function, ψ.

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