Optimally controlled quantum molecular dynamics: The effect of nonlinearities on the magnitude and multiplicity of control-field solutions

Metin Demiralp* and Herschel Rabitz
Department of Chemistry, Princeton University, Princeton, New Jersey 08544-1009
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This paper addresses the nature and multiplicity of an optimally designed electric field for controlling quantum-dynamical processes. A rather general cost functional is considered, with only mild conditions called for amongst the various operators involved. An explicit upper bound on the magnitude of the controlling electric field is attained in terms of the norms of various operators entering into the control cost functional. An earlier work employing first-order perturbation theory arguments showed that, under rather mild assumptions, a denumerably infinite number of control-field solutions exists for the optimal control problem. In the present work, it is shown that through a bound on the remainder of the nonlinear terms in the expansion, this same conclusion concerning the control-field multiplicity continues to hold.

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I. INTRODUCTION AND FORMULATION OF THE FIELD DESIGN PROBLEM

Increasing attention is being addressed toward the external control of quantum-mechanical systems [1–3]. Problems where the external control is an electromagnetic field are receiving the most attention [4–17], although other applications may also arise. In general, it is a fundamental issue to understand the degree of possible control and the practical ability to achieve such controls in the microscale world, where the laws of quantum mechanics are operative. Analogous issues are traditional areas of active pursuit in macroscale engineering, but only recently have such matters begun to be explored in the microworld. A host of topics are open for study, and the present work primarily considers the issue of a possible multiplicity of solutions to the optimal control problem in any given system. The fact that multiple solutions may exist is suggested from the observation that, typically, the control problems are specified by giving an initial condition and a desired target state, with only mild integral costs on the interior evolution of the system from the initial to final state. Thus many quantum-dynamical paths, each corresponding to a distinct (control-field) Hamiltonian, could possibly exist. In a recent paper [18], this issue was explored by the authors, taking a first-order perturbation-theory perspective, where it was found that a denumerably infinite number of solutions exist. However, the question remained open as to whether the higher-order terms in the field intensity could alter this conclusion. The present work examines this matter by placing a bound on the possible contributions of these higher-order terms. As a result, it will be shown that the detailed nature of the control solution does depend on the higher-order field nonlinearities, but the multiplicity of solutions still remains. Furthermore, in proceeding through the analysis to this conclusion, an explicit upper bound on the control-field magnitude will be obtained, in terms of the various physical variables entering the control problem.

Consideration of the role of field nonlinearities in the quantum control process will explicitly build on the prior linear formulation [18]. Thus this formulation will now be summarized, along with a precise definition of the control problem being addressed. First, the Hamiltonian of the system is defined by

$$H=H_0+\mu\mathcal{E}(t) \tag{1.1a}$$

$$=\mathcal{H}+\mu\mathcal{D}(t)\ , \tag{1.1b}$$

where H_0 is the reference Hamiltonian for the system unperturbed by the control field $\mathcal{E}(t)$. Here, this latter field is taken as coupled to the quantum-mechanical system through the dipole operator μ. The dipole and field will be treated as scalars, but their vector analogs may be just as well considered. In Eq. (1.1b), a new reference Hamiltonian

$$\mathcal{H}=H_0+\mu\tilde{\mathcal{E}} \tag{1.2}$$

has been defined in terms of a nominal background field $\tilde{\mathcal{E}}$. Correspondingly, the new effective field is shifted:

$$\mathcal{D}(t)=\mathcal{E}(t)-\tilde{\mathcal{E}}\ . \tag{1.3}$$

The nominal field $\tilde{\mathcal{E}}$ may be time dependent or time independent, and is only introduced here for convenience, since the true field $\mathcal{E}(t)$ is the one that would be generated in the laboratory. A perturbation expansion for the control problem will be written in terms of the strength of $\mathcal{D}(t)$, and altering $\tilde{\mathcal{E}}$ may improve the convergence properties of the solution. Since this introduction of $\tilde{\mathcal{E}}$ is only for this purpose, we hereafter will assume it is a constant reference field. The corresponding system Schrödinger equation will then become

$$i\hbar\frac{\partial\psi(t)}{\partial t}=[\mathcal{H}+\nu\mu\mathcal{D}(t)]\psi(t)\ , \tag{1.4}$$

where ν is introduced as an ordering parameter, to be utilized later. The optimal control problem is stated in terms of finding $\mathcal{D}(t)$, or equivalently $\mathcal{E}(t)$, such that some particular objectives are reached and penalties avoided. This perspective may be expressed as minimization of the cost functional $\mathcal{T}$,

$$\mathcal{J}=\mathcal{J}_0+\mathcal{J}_p^{(1)}+\mathcal{J}_p^{(2)}+\mathcal{J}_{c,d}+\mathcal{J}_{c,0} \tag{1.5}$$

where

$$\mathcal{J}_0=\tfrac{1}{2}[\langle\psi(T)|\hat{O}|\psi(T)\rangle-\tilde{O}]^2 , \tag{1.6a}$$

$$\mathcal{J}_p^{(1)}=\tfrac{1}{2}\int_0^T dt\; W_p(t)\langle\psi(t)|\hat{O}'|\psi(t)\rangle^2 , \quad W_p(t)>0 ,\; T\in[0,T] \tag{1.6b}$$

$$\mathcal{J}_p^{(2)}=\tfrac{1}{2}\int_0^T dt\; W_{\mathcal{E}}(t)\mathcal{E}^2(t) , \quad W_{\mathcal{E}}(t)>0 ,\; t\in[0,T] \tag{1.6c}$$

$$\mathcal{J}_{c,d}=\int_0^T dt\left\langle\lambda(t)\left|i\hbar\frac{\partial}{\partial t}-H(t)\right|\psi(t)\right\rangle+\text{c.c.} , \tag{1.6d}$$

$$\mathcal{J}_{c,0}=\eta[\langle\psi(T)|\hat{O}|\psi(T)\rangle-\tilde{O}] . \tag{1.6e}$$

In these equations, T is the target time, $\hat{O}$ is the Hermitian objective operator, $\tilde{O}$ is the target value, $\hat{O}'$ is a Hermitian penalty operator whose expectation value over the control interval is to be minimized, and the weights W_p and $W_{\mathcal{E}}$ are chosen to adjust the significance of the penalty and fluence terms $\mathcal{J}_p^{(1)}$ and $\mathcal{J}_p^{(2)}$, respectively. Note that the first and last terms in Eq. (1.5), $\mathcal{J}_0$ and $\mathcal{J}_{c,0}$, would not *both* be present in a given problem. The first of these terms represents a quadratic cost for reaching the objective, while the latter term corresponds to a constraining demand that the objective be exactly achieved. In the latter case, the Lagrange parameter η assures that this is the case, and similarly, the Lagrange function $\lambda(t)$ assures that Schrödinger's equation is satisfied. A full discussion of the meaning and significance of these terms can be found elsewhere [7–16]. The control equations are obtained by demanding that the first variation of $\mathcal{J}$ be stationary,

$$\delta\mathcal{J}=0 . \tag{1.7}$$

The resultant variational equations are

$$i\hbar\frac{\partial\psi(\nu,t)}{\partial t}=[\mathcal{H}+\nu\mu\mathcal{D}(t)]\psi(\nu,t) , \tag{1.8a}$$

$$\psi(\nu,0)=\tilde{\psi} , \tag{1.8b}$$

$$i\hbar\frac{\partial\lambda(\nu,t)}{\partial t}=[\mathcal{H}+\nu\mu\mathcal{D}(t)]\lambda(\nu,t) - W_p(t)\langle\psi(\nu,t)|\hat{O}'|\psi(\nu,t)\rangle\hat{O}'\psi(\nu,t) , \tag{1.8c}$$

$$\lambda(\nu,T)=\frac{i}{\hbar}\eta\hat{O}\psi(\nu,T) , \tag{1.8d}$$

$$\mathcal{D}(t)=\frac{2}{W_{\mathcal{E}}(t)}\text{Re}[\langle\lambda(\nu,t)|\mu|\psi(\nu,t)\rangle]-\tilde{\mathcal{E}} , \tag{1.8e}$$

$$\langle\psi(\nu,T)|\hat{O}|\psi(\nu,T)\rangle=\hat{O}+\alpha\eta , \tag{1.8f}$$

where

$$\alpha=\begin{cases}1 & \text{if } \mathcal{J}_0 \text{ is employed}\\ 0 & \text{if } \mathcal{J}_{c,0} \text{ is employed} .\end{cases} \tag{1.8g}$$

In Eq. (1.8e) Re denotes the real part. Here we have introduced the ordering parameter ν as explicitly entering the wave function $\psi(\nu,t)$ and the Lagrange multiplier function $\lambda(\nu,t)$. It is understood that the value $\nu=1$ corresponds to the physical case of interest. The variational equations naturally include Schrödinger's equation (1.8a), but most importantly, the overall set of equations (1.8) contains a third-order nonlinearity explicitly evident in Eq. (1.8c), and additionally, through the substitution of Eq. (1.8e) into Eqs. (1.8a) and (1.8c). Furthermore, for the case $\alpha=1$ in Eq. (1.8f), the final condition for $\lambda(\nu,T)$ in Eq. (1.8d) is also third-order nonlinear in terms of the wave function. Schrödinger's equation is linear from the traditional forward perspective of attaining a solution once a Hamiltonian is given, while in an optimal control framework, quantum mechanics reduces to solving a highly nonlinear problem. The origin of this nonlinearity is in the inverse nature of the control problem. The equations (1.8) that need to be solved are interestingly of a traditional nonlinear Schrödinger type [19]. The parameter η plays a very important role in establishing the multiplicity of solutions to control equations. In the case of $\alpha=1$, it is evident from Eq. (1.8f) that η provides the degree of error in reaching the control objective $\tilde{O}$. In contrast, when $\alpha=0$, then η is a Lagrange parameter, but its magnitude has a bearing on the difficulty of achieving the target $\tilde{O}$ in Eq (1.6e).

The issue of multiplicities to a quantum control problem consists of whether there are different solutions $\psi(\nu,t)$ and $\lambda(\nu,t)$ to Eqs. (1.8), and thus distinct values for the control field $\mathcal{E}(t)=\mathcal{D}(t)+\tilde{\mathcal{E}}$. In the previous work as well as here, this matter will be addressed considering the infinite-order expansions

$$\psi(\nu,t)=\sum_{j=0}^{\infty}\psi_j(t)\nu^j , \tag{1.9a}$$

$$\lambda(\nu,t)=\sum_{j=0}^{\infty}\lambda_j(t)\nu^j . \tag{1.9b}$$

Substitution of these expansions into Eqs. (1.8) and equating like powers of ν will lead to a recursive set of differential equations for the expansion functions $\psi_j(t)$ and $\lambda_j(t)$, $j=0,1,2,\ldots$ (their dependence on other coordinates, depending on the choice of representation, is implicitly understood). These equations, and their formal solution, were discussed in the earlier work on this topic [18]. For our purposes here, the only point that needs emphasis is that proper convergence behavior of Eqs. (1.9) calls for the operators μ, $\hat{O}$, and $\hat{O}'$ to be bounded. In addition, the operators $\hat{O}$ and $\hat{O}'$ are assumed to be definite (either positive or negative) in order to assure certain spectral properties to achieve the ultimate control field. Appropriate modifications can be made to guarantee that these criteria are satisfied; hereafter we will assume that μ, $\hat{O}$, and $\hat{O}'$ are properly chosen.

Addressing the issue of multiplicity of solutions reduces to consideration of Eqs. (1.8e) and (1.8f). In keeping with notation utilized previously, we define the following functions:

$$\Phi(\nu,t)=\frac{2}{W_{\mathscr{E}}}\mathrm{Re}[\langle\lambda(\nu,t)|\mu|\psi(\nu,t)\rangle]-\tilde{\mathscr{E}}\ , \quad (1.10a)$$

$$\Omega(\nu,t)=\langle\psi(\nu,t)|\hat{O}|\psi(\nu,t)\rangle\ . \quad (1.10b)$$

The sought-after field may be identified as

$$\mathscr{D}(t)=\Phi(1,t)=\frac{2}{W_{\mathscr{E}}}\mathrm{Re}[\langle\lambda(1,t)|\mu|\psi(1,t)\rangle]-\tilde{\mathscr{E}} \quad (1.11)$$

and the auxiliary function $\Omega(1,T)$ may be identified as

$$\Omega(1,T)=\tilde{O}+\alpha\eta\ . \quad (1.12)$$

Substituting the expansions in Eqs. (1.9a) and (1.9b) into Eqs. (1.10a) and (1.10b) will produce the following expansions:

$$\Phi(\nu,t)=\sum_{j=0}^{\infty}\nu^j\Phi_j(t)\ , \quad (1.13a)$$

$$\Omega(\nu,t)=\sum_{j=0}^{\infty}\nu^j\Omega_j(t)\ . \quad (1.13b)$$

Thus we have that

$$\mathscr{D}(t)=\Phi_0(t)+\Phi_1(t)+R_\Phi(t)\ , \quad (1.14a)$$

where

$$R_\Phi(t)=\sum_{j=2}^{\infty}\Phi_j(t) \quad (1.14b)$$

and similarly

$$\Omega_0(T)+\Omega_1(T)+R_\Omega(T)=\tilde{O}+\alpha\eta\ , \quad (1.15a)$$

where

$$R_\Omega(T)=\sum_{j=2}^{\infty}\Omega_j(T)\ . \quad (1.15b)$$

In earlier work [18], Eqs. (1.14a) and (1.15a) were referred to as the field and spectral equations, respectively. In the latter work, the remainder terms R_Φ and R_Ω were ignored, and the resultant truncated equations are linear with respect to the unknown field. In particular, the field $\mathscr{D}_L(t)$ arising from truncation of Eq. (1.14a) was shown to be expressible in terms of an expansion in a special set of time-independent eigenfunctions $e_k(t)$,

$$\mathscr{D}_L(t)=\eta\sum_{k=1}^{\infty}\frac{(e_k,u_1)}{\eta_k-\eta}e_k(t)\ , \quad (1.16)$$

and the reader is referred to the prior reference for a precise definition of the inner product (e_k,u_1) and the parameters (eigenvalues) η_k entering into Eq. (1.16). Similarly, the truncated form of Eq. (1.15a), without the remainder term R_Ω was shown to become

$$\sum_{k=1}^{\infty}\frac{(e_k,u_1)^2}{\eta_k-\eta}=\alpha+\{\hat{O}-\langle\tilde{\psi}|Q(T)|\tilde{\psi}\rangle\}\frac{1}{\eta}\ , \quad (1.17)$$

where

$$Q(T)=\exp\left[\frac{i}{\hbar}T\mathscr{H}\right]\hat{O}\exp\left[-\frac{i}{\hbar}T\mathscr{H}\right]\ . \quad (1.18)$$

Thus the field in Eq. (1.16) depends on the free parameter η, which is determined from solving Eq. (1.17) for this parameter. A simple analysis showed that Eq. (1.17) has a denumerably infinite number of solutions $\eta^{(l)}$, $l=1,2,\ldots$. These roots, in turn, specify a denumerably infinite number of control fields in Eq. (1.16). The significance of this result is that many of these solutions may have physically acceptable qualities.

The purpose of the present paper is to investigate the simultaneous solutions of Eqs. (1.14a) and (1.15a), retaining the remainder terms R_Φ and R_Ω. These latter terms functionally depend on the unknown sought-after field $\mathscr{D}(t)$ to second and all higher orders. Thus the identification of the solution multiplicity of these equations appears to be an extremely difficult task. However, we will show in the subsequent sections that an explicit upper bound may be placed on the magnitude of the remainder terms in these equations, and from the behavior of these upper bounds we may once again draw simple conclusions concerning the multiplicity of optimal control solutions. In the process of obtaining these bounds and the final multiplicity results, an interesting expression will be obtained for an upper bound to the sought-after field.

The remainder of this paper is organized as follows. In Sec. II, various intermediary bounding expressions will be derived; in addition, this section will also present the bound for the control field. In Sec. III, an explicit bound will be obtained for R_Φ and R_Ω, and from these bounds, arguments will be presented on the multiplicity of solutions to the control equations. Finally, Sec. IV will present some summarizing comments.

II. INEQUALITIES AND UPPER BOUNDS ON THE CONTROL FIELD AND OBJECTIVES

Following the analysis in the previous paper [18], it may be readily seen that Eqs. (1.14a) and (1.15a) are nonlinear (to infinite order) integral equations in terms of the control field $\mathscr{D}(t)$. Our purpose here is not to explicitly solve these equations, but rather to explore some properties of their solutions. The approach taken here is to identify upper bounds on a variety of intermediate expectation values and inner products necessary to prove the multiplicity results in Sec. III. In the process, an explicit upper bound will be obtained for the field strength itself.

A. Upper bound on the optimal control field

If we now denote the absolute value bound for the molecular dipole function by μ_B, then together with the normalization condition

$$\langle\psi(\nu,t)|\psi(\nu,t)\rangle=1 \quad (2.1)$$

we have

$$|\langle\psi(\nu,t)|\mu^k|\psi(\nu,t)\rangle|<\mu_B^k\ ,\quad k\geq 1\ . \quad (2.2)$$

We desire to find an upper bound $\mathscr{D}_B(t)$ for the deviation in the field amplitude $\mathscr{D}(t)$ from its nominal value $\tilde{\mathscr{E}}$. To this end, all we have to do is find a bound for the norm of the Lagrange multiplier $\lambda(\nu,t)$ (or costate function) that satisfies the backward evolutionary equations (1.8c) and (1.8d). By employing these equations and their complex conjugated analogs, together with the self-adjointness of the operators involved, we can conclude that

$$\frac{\partial\langle\lambda(\nu,t)|\lambda(\nu,t)\rangle}{\partial t}=-\frac{2}{\hbar}W_p(t)\langle(\nu,t)|\hat{O}'|\psi(\nu,t)\rangle \times\text{Im}[\langle\lambda(\nu,t)|\hat{O}'|\psi(\nu,t)\rangle] , \quad (2.3)$$

$$\langle\lambda(\nu,T)|\lambda(\nu,T)\rangle=\frac{\eta^2}{\hbar^2}\langle\psi(\nu,T)|\hat{O}^2|\psi(\nu,T)\rangle . \quad (2.4)$$

Here Im denotes the imaginary part. The integration of Eq. (2.3) over the interval $[t,T]$ and use of the boundedness of the operators appearing here, together with the unit normalization of $\psi(\nu,t)$, enables us to arrive at the following conclusion after repeated use of the Cauchy-Schwartz inequality for the inner products:

$$\langle\lambda(\nu,t)|\lambda(\nu,t)\rangle<\frac{\eta^2||\hat{O}||^2}{\hbar^2}+\frac{2||\hat{O}'||^2}{\hbar}\left[\int_0^T d\tau\, W_p^2(\tau)\right]^{1/2}\left[\int_0^T d\tau\langle\lambda(\nu,\tau)|\lambda(\nu,\tau)\rangle\right]^{1/2} . \quad (2.5)$$

The integration of this equation with respect to t from 0 to T produces the following inequality:

$$\mathcal{A}\mathcal{X}^2+\mathcal{B}\mathcal{X}+\mathcal{C}<0 , \quad (2.6a)$$

$$\mathcal{A}\equiv1 , \quad \mathcal{B}\equiv-\frac{2T||\hat{O}'||^2}{\hbar}\left[\int_0^T d\tau\, W_p^2(\tau)\right]^{1/2} , \quad (2.6b)$$

$$\mathcal{C}\equiv\frac{\eta^2||\hat{O}||^2T}{\hbar^2} ,$$

$$\mathcal{X}\equiv\left[\int_0^T d\tau\langle\lambda(\nu,\tau)|\lambda(\nu,\tau)\rangle\right]^{1/2} . \quad (2.6c)$$

The solution of the inequality in Eq. (2.6a) shows that $\mathcal{X}$ is bounded from below by 0 and bounded above by the largest root of the trinomial given in Eqs. (2.6),

$$\mathcal{X}=\left[\int_0^T dt\langle\lambda(\nu,t)|\lambda(\nu,t)]\right]^{1/2} <\frac{T||\hat{O}'||^2}{\hbar}\left[\int_0^T dt\, W_p^2(t)\right]^{1/2} +\frac{T}{\hbar}\left[||\hat{O}'||^4\int_0^T dt\, W_p^2(t)+\frac{\eta^2||\hat{O}||^2}{T}\right]^{1/2} . \quad (2.7)$$

This result can be put into a more amenable form by making use of the following inequality, which holds for all positive values of A and B:

$$(A+B)^{1/2}<\sqrt{A}+\frac{B}{2\sqrt{A}} , \quad (2.8)$$

where the quantities A and B are identified as

$$A=||\hat{O}'||^4\int_0^T dt\, W_p^2(t) , \quad B=\frac{\eta^2||\hat{O}||^2}{T} . \quad (2.9)$$

The inequality in Eq. (2.8) enables us to expand the second term on the right-hand side of Eq. (2.7). The result may then be substituted into the right-hand side of Eq. (2.5) to obtain the bound.

$$\langle\lambda(\nu,t)|\lambda(\nu,t)\rangle<\frac{2\eta^2||\hat{O}||^2}{\hbar}+\frac{4T||\hat{O}'||^4}{\hbar}\int_0^T dt\, W_p^2(t) . \quad (2.10)$$

Considering Eq. (1.8e) makes it possible to derive an upper bound for the control field by using the inequalities that are already derived in this section,

$$|\mathcal{D}(t)|<\mathcal{D}_B=\frac{2\mu_b}{\hbar W_{\mathcal{E}}(t)}\left[2\eta^2||\hat{O}||^2+4T||\hat{O}'||^4 \times\int_0^T dt\, W_p^2(t)\right]^{1/2}+|\tilde{\mathcal{E}}| . \quad (2.11)$$

It is interesting to examine the dependence of this bound upon the physical variables involved. The inverse dependence on the weight $W_{\mathcal{E}}(t)$ is reasonable, as a lowering of its value would allow for more effective control, through an increase of the field intensity. Recognizing that η is a deviation from the target state (or a measure of how difficult it is to achieve the target exactly in the constrained case), it is reasonable to expect that the field bound should grow with this variable. Similarly, an increase in the norm of the target operator $||\hat{O}||$ will naturally call for an increase in the control-field intensity. Similar arguments apply to an increase in the penalty operator norm $||\hat{O}'||$ and its associated weight $W_p(t)$. It is also interesting that, in the latter case, there is growth to the control field with T, due to the accumulated integral nature of the penalty term in the cost functional. There is also a hidden dependence on T in η, as η, being the target error term, is expected to decrease to an asymptotic value as T increases. Finally, the proportionality of the result to the dipole moment norm μ_B may seem surprising, but it arises since the fluence term in the cost functional is just with respect to the field intensity and not the dipole interaction energy. The conservative nature of the bound in Eq. (2.11) is not known, but it provides interesting insight into the factors controlling the field intensity.

B. Inequalities, expectation values, and inner products

The norm in Eq. (2.11), along with several other inequalities associated with the first- and second-order partial derivatives of $\psi(\nu,t)$ and $\lambda(\nu,t)$, with respect to ν, will be needed in the next section to explore the multiplicity of solutions for the optimal control equations. Let $\psi_\nu(\nu,t)$ and $\psi_{\nu\nu}(\nu,t)$ denote the first- and second-order partial derivatives of $\psi(\nu,t)$ with respect to ν. Then, $\psi_\nu(\nu,t)$ satisfies the following evolution equation:

$$i\hbar\frac{\partial\psi_\nu(\nu,t)}{\partial t}=[\mathcal{H}+\nu\mu\mathcal{D}(t)]\psi_\nu(\nu,t)+\mu\mathcal{D}(t)\psi(\nu,t) , \tag{2.12a}$$

$$\psi_\nu(\nu,0)=0 . \tag{2.12b}$$

By using this equation and its complex conjugate, we can obtain a differential equation for the norm of $\psi_\nu(\nu,t)$. Its integration over time from 0 to t gives the following equation:

$$\langle\psi_\nu(\nu,t)|\psi_\nu(\nu,t)\rangle =\frac{2}{\hbar}\int_0^t d\tau\mathcal{D}(\tau)\mathrm{Im}[\langle\psi_\nu(\nu,\tau)|\mu|\psi(\nu,\tau)\rangle] . \tag{2.13}$$

By using the Cauchy-Schwartz inequality for the kernel of the integral on the right-hand side, we can obtain the following inequality:

$$\langle\psi_\nu(\nu,t)|\psi_\nu(\nu,t)\rangle<\frac{2\mu_B}{\hbar}\left[\int_0^t d\tau\mathcal{D}_B^2(\tau)\right]^{1/2}\times\left[\int_0^T d\tau\langle\psi_\nu(\nu,\tau)|\mu|\psi(\nu,\tau)\rangle\right]^{1/2} . \tag{2.14}$$

The integration of this inequality over $[0,T]$ on t reveals the upper bound for the integral of the norm of $\psi_\nu(\nu,\tau)$, and utilization of this result in Eq. (2.13) enables us to conclude

$$\langle\psi_\nu(\nu,t)|\psi_\nu(\nu,t)\rangle<\frac{4T\mu_B^2}{\hbar^2}\int_0^T d\tau\mathcal{D}_B^2(\tau) \tag{2.15}$$

and therefore

$$\langle\psi_\nu(\nu,t)|\mu^k|\psi_\nu(\nu,t)\rangle<\frac{4T\mu_B^{k+2}}{\hbar^2}\int_0^T d\tau\mathcal{D}_B^2(\tau) , \quad k\geq 1 . \tag{2.16}$$

After a similar analysis, we can write the following results for $\psi_{\nu\nu}(\nu,t)$:

$$\langle\psi_{\nu\nu}(\nu,t)|\psi_{\nu\nu}(\nu,t)\rangle<\frac{64T^2\mu_B^4}{\hbar^4}\int_0^T d\tau\mathcal{D}_B^2(\tau) , \tag{2.17}$$

$$\langle\psi_{\nu\nu}(\nu,t)|\nu^k|\psi_{\nu\nu}(\nu,t)\rangle<\frac{64T^2\mu_B^{k+4}}{\hbar^4}\int_0^T d\tau\mathcal{D}_B^2(\tau) , \quad k\geq 1 . \tag{2.18}$$

Similar steps can be followed to obtain the norms of the first- and second-order partial derivatives of $\lambda(\nu,t)$, with respect to ν. We only report the results here:

$$\langle\lambda_\nu(\nu,t)|\lambda_\nu(\nu,t)\rangle<\frac{8T\mu_B^2}{\hbar^4}\left[\int_0^T d\tau\mathcal{D}_B^2(\tau)\right]\left[\eta^2||\hat{O}||^2+18T||\hat{O}'||^4\int_0^T d\tau\, W_p^2(\tau)\right] , \tag{2.19}$$

$$\langle\lambda_{\nu\nu}(\nu,t)|\lambda_{\nu\nu}(\nu,t)\rangle<\frac{128T^2\mu_B^4}{\hbar^6}\left[\int_0^T d\tau\mathcal{D}_B^2(\tau)\right]^2\left[\eta^2||\hat{O}||^2+72T||\hat{O}'||^4\int_0^T d\tau\, W_p^2(\tau)\right] . \tag{2.20}$$

These various inequalities will now be utilized in the next section.

III. BOUNDS FOR THE NONLINEAR TERMS IN THE FIELD AND THE SPECTRAL EQUATIONS AND OPTIMAL CONTROL MULTIPLICITY

Now we are sufficiently equipped to investigate the effects of the nonlinear terms in field and spectral equations (1.13a) and (1.13b) upon the multiplicity of control-field solutions. In these latter equations, $\Phi_j(t)$ and $\Omega_j(T)$ are jth-degree homogeneous functionals of $\mathcal{D}(t)$. Hence, there is well ordering in terms of the deviation in the field amplitude from its nominal value.

As we did in the earlier paper [18] and summarized in Sec. I, the field equation (1.14a) and the spectral equation (1.15a) can be linearized by omitting $\Phi_j(t)$ and $\Omega_j(t)$ when the index j is greater than 1. This linearization led us to obtain a weighted eigenvalue problem involving integral operators for the field equation, and a multibranch algebraic equation with an infinite number of vertical asymptotes for the spectral equation. The issue is now the determination of the effects of the nonlinear terms upon the structure and solution of these linearized equations. Let us consider the well-known Taylor identity for a twice-differentiable function on a closed interval as follows:

$$f(x)=f(0)+xf'(0)+\int_0^x dy(x-y)f''(y) . \tag{3.1}$$

If the interval on x is [0,1] then we can further write

$$f(1)=f(0)+f'(0)+\int_0^1 dy(1-y)f''(y) , \tag{3.2}$$

which enables us to obtain the following analogs for $\Phi(\nu,t)$ and $\Omega(\nu,t)$:

$$\Phi(1,t)=\Phi(0,t)+\Phi_\nu(0,t)+\int_0^1 d\nu(1-\nu)\Phi_{\nu\nu}(\nu,t) , \tag{3.3}$$

$$\Omega(1,t)=\Omega(0,t)+\Omega_\nu(0,t)+\int_0^1 d\nu(1-\nu)\Omega_{\nu\nu}(\nu,t) , \tag{3.4}$$

where the subscripts ν and $\nu\nu$ mean the first- and second-order partial differentiation with respect to ν, respectively. From a comparison of Eqs. (3.3) and (3.4) with Eqs. (1.13a) and (1.13b), we see that $\Phi_0(t)=\Phi(0,t)$, $\Phi_1(t)=\Phi_\nu(0,t)$, $\Omega_0(T)=\Omega(0,T)$, and $\Omega_1(T)=\Omega_\nu(0,T)$.

These equations are linear in $\mathcal{D}(t)$ except the right-hand side integrals, which may be identified from Eqs. (1.14b) and (1.15b) as

$$R_\Phi(t)=\int_0^1 d\nu(1-\nu)\Phi_{\nu\nu}(\nu,t)\ , \tag{3.5}$$

$$R_\Omega(t)=\int_0^1 d\nu(1-\nu)\Omega_{\nu\nu}(\nu,t)\ . \tag{3.6}$$

Hence all nonlinear contributions to the field and to the spectral equations are gathered in these integrals. Once we find appropriate bounds for them, we can analyze the effects of the nonlinearities on the solutions of the linearized field and spectral equations.

Now we can write the following inequality for the kernel of the integral in Eq. (3.5) from the definition in Eq. (1.10a):

$$|\Phi_{\nu\nu}(\nu,t)|<\frac{2}{W_{\mathcal{E}}(t)}|\langle\lambda_{\nu\nu}(\nu,t)|\mu|\psi(\nu,t)\rangle| +\frac{2}{W_{\mathcal{E}}(t)}|\langle\lambda_{\nu}(\nu,t)|\mu|\psi_{\nu}(\nu,t)\rangle| +\frac{2}{W_{\mathcal{E}}(t)}|\langle\lambda(\nu,t)|\mu|\psi_{\nu\nu}(\nu,t)\rangle|\ . \tag{3.7}$$

By using the boundedness of μ and the Cauchy-Schwartz inequality for inner products, we can obtain the following result, after some intermediate algebra:

$$|\Phi_{\nu\nu}(\nu,t)|<\frac{40\sqrt{2}T\mu_B^3}{\hbar^3 W_{\mathcal{E}}(t)}\left[\eta^2||\hat{O}||^2+72T||\hat{O}'||^4\int_0^T d\tau\, W_p^2(\tau)\right]^{1/2}\int_0^T d\tau\,\mathcal{D}_B^2(\tau)\ . \tag{3.8}$$

Since the right-hand side of this equation does not depend on ν, we can write

$$\left|\int_0^1 d\nu(1-\nu)\Phi_{\nu\nu}(\nu,t)\right|<\frac{20\sqrt{2}T\mu_B^3}{\hbar^3 W_{\mathcal{E}}(t)}\left[\eta^2||\hat{O}||^2+72T||\hat{O}'||^4\int_0^T d\tau\, W_p^2(\tau)\right]^{1/2}\int_0^T d\tau\,\mathcal{D}_B^2(\tau)\ . \tag{3.9}$$

Combining Eq. (3.9) with the bound $\mathcal{D}_B(t)$ in Eq. (2.11) provides an explicit bound for the remainder term $R_\Phi(t)$ in Eq. (1.14a). Since all nonlinearities in the field equation are represented by a single bounded functional of the physical variables, we can simply conclude that there is no irregularity or drastic changes in the mathematical structure of the field equation when the nonlinear terms are included.

Now, we can proceed in a similar way for the spectral equation and obtain the following result:

$$|\Omega_{\nu\nu}(\nu,t)|<\frac{24T\mu_B^2||\hat{O}||}{\hbar^2}\int_0^T d\tau\,\mathcal{D}_B^2(\tau)\ , \tag{3.10}$$

which means that

$$\left|\int_0^1 d\nu(1-\nu)\Omega_{\nu\nu}(\nu,t)\right|<\frac{12T\mu_B^2||\hat{O}||}{\hbar^2}\int_0^T d\tau\,\mathcal{D}_B^2(\tau)\ . \tag{3.11}$$

This result, when combined with the bound for $\mathcal{D}_B(t)$ in Eq. (2.11), produces an explicit bound for $R_\Omega(T)$ entering into Eq. (1.15a). Since $R_\Omega(T)$ is bounded, there is no effect on the structure of the eigenvalue problem associated with the linearized equations, except for a change in the values of η satisfying Eq. (1.15a). Thus, although the particular nature of each field solution to the optimal control problem may change due to system nonlinearities, the basic conclusion of there being a denumerably infinite number of solutions still remains.

IV. CONCLUDING REMARKS

This paper presented a general proof that there will be a denumerably infinite number of solutions to a quantum-mechanical optimal control problem. Furthermore, an explicit upper bound was obtained on the optimal control field in terms of the system physical variables. These conclusions were made under some rather mild assumptions. First, the operators μ, $\hat{O}$, and $\hat{O}'$ were assumed to be bounded, and second, the operators $\hat{O}$ and $\hat{O}'$ were assumed to be definite. A violation of these conditions can bring into question the convergence properties of the wave function and Lagrange multiplier function explicitly utilized in the proof. Perhaps more importantly, these results are based on the chosen form of the cost functional in Eqs. (1.5) and (1.6). Although this latter form is quite general, other cost functionals, including simpler ones, might also be considered. It would be valuable to explore this matter further to see whether different conclusions might be reached, concerning optimal field multiplicity.

The presence of optimal field multiplicity is potentially quite significant for quantum-mechanical control. First, the solutions with smaller values of the error index η obtained from solving Eq. (1.5a) correspond to results where the objective expectation value is closer to its target value in Eq. (1.8f) (or is more easily achieved in the case of constrained target value). However, in practice, many other solutions corresponding to different η values may be quite acceptable, as, in fact, it was the minimization of the total cost functional $\mathcal{T}$ which was defined as the truly interesting objective in the quantum-mechanical control problem. The possibility of there being many multiple minima of acceptable quality merely opens up the prospect of adding further auxiliary criteria, or costs, to the optimizing functional, in order to obtain solutions meeting further physical demands. One cautionary point is that the presence of multiple solutions under certain circumstances may cause numerical difficulties in searching through the control function space for solutions. This is an algorithmic matter, which deserves close attention.

Finally, although this paper and its earlier companion [18] aimed at a general analysis of the quantum-mechanical optimal control problem, numerical methods based on perturbation theory may nonetheless also be of practical significance. Clearly, the place to start in this regard is the lowest order of perturbation theory in the earlier paper. In this case, the multiplicity of solutions may be explored without resorting to complicated iterative optimization—only a one-dimensional root search for the characteristic error parameter η needs to be performed. Even if the results are only qualitatively correct in the lowest-order perturbation formulation, they should nonetheless be interesting for their physical content on the type and variety of solutions.

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

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