Assessing optimality and robustness of control over quantum dynamics

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(Received 6 October 1997)

This work presents a general framework for assessing the quality and robustness of control over quantum dynamics induced by an optical field $\mathcal{E}(t)$. The control process is expressed in terms of a cost functional, including the physical objectives, penalties, and constraints. The first variations of such cost functionals have traditionally been utilized to create designs for the controlling electric fields. Here, the second variation of the cost functional is analyzed to explore (i) whether such solutions are locally optimal, and (ii) their degree of robustness. Both issues may be assessed from the eigenvalues of the stability operator $\mathcal{S}$ whose kernel $K(t,\tau)$ is related to $\delta\mathcal{E}(t)/\delta\mathcal{E}(\tau)|_c$ for $0<t,\ \tau\leq T$, where T is the target control time. Here c denotes the constraint that the field satisfies the optimal control dynamical equations. The eigenvalues σ of $\mathcal{S}$ satisfying $\sigma<1$ assure local optimality of the control solution, with $\sigma=1$ being the critical value separating optimal solutions from false solutions (i.e., those with negative second variational curvature of the cost functional). In turn, the maximally robust control solutions with the least sensitivity to field errors also correspond to $\sigma=1$. Thus, sufficiently high sensitivity of the field at one time t to the field at another time τ (i.e., $\sigma>1$) will lead to a loss of local optimality. An expression is obtained for a bound on the stability operator, and this result is employed to qualitatively analyze control behavior. From this bound, the inclusion of an auxiliary operator (i.e., other than the target operator) is shown to act as a stabilizer of the control process. It is also shown that robust solutions are expected to exist in both the strong- and weak-field regimes. [S1050-2947(98)07203-5]

PACS number(s): 32.90.+a

I. INTRODUCTION

In recent years, there has been considerable activity in the domain of designing controls to actively manipulate quantum dynamics phenomena [1–3]. There are many potential applications in chemistry, physics, and nanoscale engineering. Most attention has been focused on designing optical electric fields for dynamical manipulation, and the design techniques have ranged from perturbation theory [2] to the exploitation of fully nonlinear techniques [4]. It has been shown [5] that the most general approach is through optimal control theory [6], and numerous theoretical efforts have explored control designs for manipulating various rotational, vibrational, and electronic processes. Throughout this research, the primary focus has been on obtaining reliable control field designs, and little attention has been paid to the robustness of these designs. The present paper considers general criteria for establishing robustness and the optimality of the control solutions. Control field design within optimal control theory [1,3,4,7,8] is based on first prescribing a physically motivated cost functional $\mathcal{J}$, which contains all of the information about the physical objectives and any penalties or constraints imposed on the dynamical evolution. It is generally understood that there can be multiple extrema $\delta\mathcal{J}/\delta\mathcal{E}(t)=0$, with respect to the field, for any particular control problem. The physically acceptable solutions correspond to a minimization of $\mathcal{J}$, and the first variation criterion $\delta\mathcal{J}/\delta\mathcal{E}(t)=0$ does not guarantee whether the solution is a local minimum or maximum of $\mathcal{J}$. This circumstance can only be assessed by considering the second variation $\delta^2\mathcal{J}/\delta\mathcal{E}(t)\delta\mathcal{E}(\tau)$, and determining its positive or negative definite character at each solution determined from the first variational equations. Even if solutions are determined to be physically acceptable as minima, it is also highly desirable that they be robust to arbitrary incremental variations $\delta\mathcal{E}(t)$ in the control field, as might arise due to errors or uncertainties in the laboratory. In this context, robustness corresponds to a solution associated with minimal positive curvature of the cost functional. Assessment of these matters involves functional analysis, but we may qualitatively understand the situation by considering the reduced problem of a single control parameter α for illustration. This situation is depicted in Fig. 1. All of the critical points α_i, $i=1,\ldots,4$ correspond to solutions of the first variational equations $\partial\mathcal{J}/\partial\alpha_i=0$, but the cases α_1 and α_3 are not acceptable, as they do not minimize $\mathcal{J}$. The solutions α_2 and α_4 are both locally optimal, and the case of α_2 gives the best solution, in the sense that $\mathcal{J}(\alpha_2)<\mathcal{J}(\alpha_4)$. However, from a robustness point of view, the solution α_4 is better, as $\partial^2\mathcal{J}/\partial\alpha_4^2<\partial^2\mathcal{J}/\partial\alpha_2^2$. The solution at α_4 is more robust than that at α_2, since, for a given arbitrary small variation δ of α, we find that $|\mathcal{J}(\alpha_4+\delta)-\mathcal{J}(\alpha_4)|<|\mathcal{J}(\alpha_2+\delta)-\mathcal{J}(\alpha_2)|$. In general, we may identify the best control solution as the one that simultaneously minimizes $\mathcal{J}$ while having the smallest curvature. As is found in Fig. 1, it may happen that a tradeoff exists between the absolute quality of the achieved solution and its robustness. This is a problem that is only likely to be identifiable on a problem-by-problem basis, and the present paper is concerned with more general considerations.

In this work, we show that eigenvalues of the stability operator $\mathcal{S}$, whose kernel $\mathcal{K}(t,\tau)$ is related to the dynamically constrained (c) functional derivative $\delta\mathcal{E}(t)/\delta\mathcal{E}(\tau)|_c$ for

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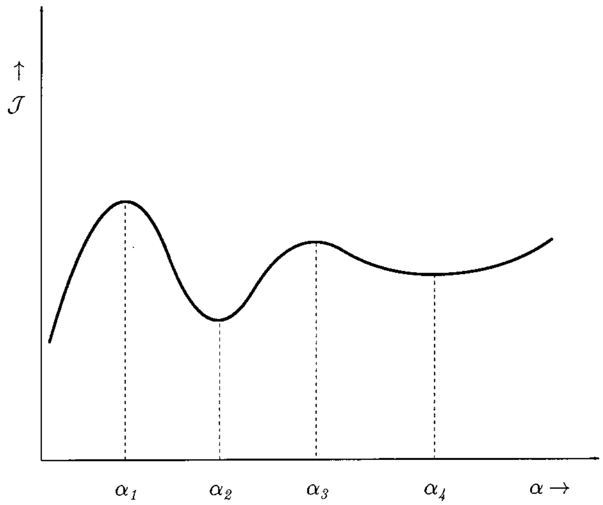

FIG. 1. A cost functional is schematically shown to depend on an optimal control parameter α as an illustration of optimality and robustness circumstances. Each of the points α_i, $i=1,\ldots,4$ satisfies $\delta\mathcal{J}=0$; however, only the cases α_2 and α_4 are physically acceptable locally optimal solutions. One of these two cases α_2 gives better control, while α_4 gives better robustness. The circumstances for all four critical values of α have a functional extension to the general control field $\mathcal{E}(t)$.

$0<t$, $\tau\leqslant T$, dictates both the optimality and robustness of potential control solutions for manipulating quantum dynamics. A formal expression for this operator will be identified, with bounds placed on its spectrum. Some qualitative conclusions on the nature of robustness will be drawn from this bounding relationship.

The paper is organized as follows: Section II presents the formal analysis leading to the expression for the stability operator; Sec. III places a bound on this operator which permits a qualitative robustness and optimality analysis. Some brief concluding remarks are presented in Sec. IV.

II. IDENTIFICATION OF THE STABILITY OPERATOR

Consider quantum motion under the influence of an external field $\mathcal{E}(t)$ described by the following Hamiltonian:

$$H=H_0+\mathcal{E}(t)\mathcal{D}, \tag{2.1}$$

where H_0, $\mathcal{D}$, and $\mathcal{E}(t)$ respectively denote the Hamiltonian for the free motion, the dipole operator projected along the direction of the external field, and the amplitude of the external field. The following cost functional prescribes the optimal control of this system [1,3,4]:

$$\mathcal{J}=\mathcal{J}_o+\mathcal{J}_p^{(1)}+\mathcal{J}_p^{(2)}+\mathcal{J}_{c,d}. \tag{2.2}$$

Here, $\mathcal{J}_o$ is the objective term that measures the difference between the expectation value of a given objective operator $\hat{O}$ and its target value $\tilde{O}$,

$$\mathcal{J}_o=\tfrac{1}{2}[\langle\psi(T)|\hat{O}|\psi(T)\rangle-\tilde{O}]^2. \tag{2.3}$$

Control through application of $\mathcal{E}(t)$ is affected over the time interval $0<t\leqslant T$, with T being the target time.

The penalty term $\mathcal{J}_p^{(1)}$ aims to suppress the expectation value of a given ''undesirable'' operator $\hat{O}'$,

$$\mathcal{J}_p^{(1)}=\tfrac{1}{2}\int_0^T dt\; W_p(t)\langle\psi(t)|\hat{O}'|\psi(t)\rangle^2,$$

$$W_p(t)>0,\quad t\in[0,T]. \tag{2.4}$$

The second penalty term allows for the possibility of minimizing the field fluence,

$$\mathcal{J}_p^{(2)}=\tfrac{1}{2}\int_0^T dt\; W_{\mathcal{E}}(t)\mathcal{E}(t)^2,$$

$$W_{\mathcal{E}}(t)>0,\quad t\in[0,T]. \tag{2.5}$$

The term denoted by $\mathcal{J}_{c,d}$ includes the dynamical constraint that Schrödinger's equation must be satisfied. This is assured through the introduction of a Lagrange multiplier λ, as given by

$$\mathcal{J}_{c,d}=2\,\mathrm{Re}\left(\int_0^T dt\left\langle\lambda(t)\left|i\hbar\frac{\partial}{\partial t}-H\right|\psi(t)\right\rangle\right). \tag{2.6}$$

Additional terms may be added to the cost functional in Eq. (2.2), but the present form covers most applications.

A control solution is attained by considering the first variation of the cost functional

$$\delta\mathcal{J}=\int_0^T dt\; W_{\mathcal{E}}(t)\,\delta\mathcal{E}(t)\mathcal{E}(t)$$

$$-2\int_0^T dt\;\delta\mathcal{E}(t)\mathrm{Re}\langle\lambda(t)|\mathcal{D}|\psi(t)\rangle. \tag{2.7}$$

Equation (2.7) has already exploited the vanishing of the first variations with respect to $|\psi(t)\rangle$ and $|\lambda(t)\rangle$, to respectively give Schrödinger's equation where

$$i\hbar\frac{\partial|\bar{\psi}(t)\rangle}{\partial t}=[H_0+\bar{\mathcal{E}}(t)\mathcal{D}]|\bar{\psi}(t)\rangle, \tag{2.8a}$$

$$|\bar{\psi}(0)\rangle=|f\rangle, \tag{2.8b}$$

where $|f\rangle$ is the initial state, and the equation for the Lagrange multiplier

$$i\hbar\frac{\partial|\bar{\lambda}(t)\rangle}{\partial t}=[H_0+\bar{\mathcal{E}}(t)\mathcal{D}]|\bar{\lambda}(t)\rangle-W_p(t)$$

$$\times\langle\bar{\psi}(t)|\hat{O}'|\bar{\psi}(t)\rangle\hat{O}'\bar{\psi}(t), \tag{2.9a}$$

$$|\bar{\lambda}(T)\rangle=\frac{i}{\hbar}\,\eta\hat{O}|\bar{\psi}(T)\rangle, \tag{2.9b}$$

$$\eta=\langle\bar{\psi}(T)|\hat{O}|\bar{\psi}(T)\rangle-\tilde{O}. \tag{2.9c}$$

Here the overbars label the functions that satisfy the first variation of the cost functional as zero. Finally, considering the variation with respect to the field in Eq. (2.7), we obtain

$$\overline{\mathcal{E}}(t)=\frac{2}{W_{\mathcal{E}}(t)}\,\mathrm{Re}[\langle\overline{\lambda}(t)|\mathcal{D}|\overline{\psi}(t)\rangle]. \tag{2.10}$$

Equations (2.8)–(2.10) will typically have multiple solutions [7]. Without further analysis one can not be sure that the solutions truly minimize the cost functional $\mathcal{J}$ and whether they are robust to variations in the control field. These considerations can be investigated by examining the second-order variation of the cost functional

$$\begin{aligned}\overline{\delta^2\mathcal{J}}=&\int_0^T dt\; W_{\mathcal{E}}(t)\,\delta\mathcal{E}(t)^2\\&-2\int_0^T dt\;\;\delta\mathcal{E}(t)\mathrm{Re}[\langle\overline{\delta\lambda}(t)|\mathcal{D}|\psi(t)\rangle\\&+\langle\lambda(t)|\mathcal{D}|\overline{\delta\psi}(t)\rangle],\end{aligned} \tag{2.11}$$

where $|\overline{\delta\lambda}\rangle$ and $|\overline{\delta\psi}\rangle$, respectively, stand for the first variations of the wave function and the Lagrange multiplier function, which are evaluated at the optimal values of the wave function, $|\overline{\psi}(t)\rangle$, Lagrange multiplier function, $|\overline{\lambda}(t)\rangle$, and the field amplitude $\overline{\mathcal{E}}(t)$. To utilize Eq. (2.11) we need to evaluate $|\overline{\delta\lambda}(t)\rangle$ and $|\overline{\delta\psi}(t)\rangle$ in terms of $|\overline{\psi}(t)\rangle$, $|\overline{\lambda}(t)\rangle$, and $\overline{\mathcal{E}}(t)$. For this purpose, we will employ Eqs. (2.8a) and (2.8b) and (2.9a) and (2.9b). The Eqs. (2.8a) and (2.8b) describe the forward quantum dynamics of the optimally controlled system under consideration. Although they are written for specific optimal values of the field amplitude $\overline{\mathcal{E}}(t)$, they remain valid for any arbitrary $\mathcal{E}(t)$. If we denote the corresponding wave function by $|\psi(t)\rangle$ in this general case, then we can rewrite the Eqs. (2.8a) and (2.8b) by removing the overbars from the relevant entities as follows:

$$i\hbar\,\frac{\partial|\psi(t)\rangle}{\partial t}=[H_0+\mathcal{E}(t)\mathcal{D}]|\psi(t)\rangle, \tag{2.12a}$$

$$|\psi(0)\rangle=|f\rangle. \tag{2.12b}$$

Now we can take the first variation of this dynamical equation to arrive at the following equation after insertion of the optimal values of each entry:

$$i\hbar\,\frac{\partial|\overline{\delta\psi}(t)\rangle}{\partial t}=[H_0+\overline{\mathcal{E}}(t)\mathcal{D}]|\overline{\delta\psi}(t)\rangle+\delta\mathcal{E}(t)\mathcal{D}|\overline{\psi}(t)\rangle, \tag{2.13a}$$

$$|\overline{\delta\psi}(0)\rangle=0, \tag{2.13b}$$

where $\delta\mathcal{E}(t)$ is considered completely arbitrary.

A similar derivation also holds for evaluation of $|\overline{\delta\lambda}(t)\rangle$ to yield

$$\begin{aligned}i\hbar\,\frac{\partial|\overline{\delta\lambda}(t)\rangle}{\partial t}=&[H_0+\overline{\mathcal{E}}(t)\mathcal{D}]|\overline{\delta\lambda}(t)\rangle+\delta\mathcal{E}(t)\mathcal{D}|\overline{\lambda}(t)\rangle-W_p(t)\\&\times\langle\overline{\psi}(t)|\hat{O}'|\overline{\psi}(t)\rangle\hat{O}'|\overline{\delta\psi}(t)\rangle\\&-2W_p(t)\mathrm{Re}[\langle\overline{\delta\psi}(t)|\hat{O}'|\overline{\psi}(t)\rangle]\hat{O}'|\overline{\psi}(t)\rangle,\end{aligned} \tag{2.14a}$$

$$\begin{aligned}|\overline{\delta\lambda}(T)\rangle=&\frac{2i}{\hbar}\,\mathrm{Re}[\langle\overline{\delta\psi}(T)|\hat{O}|\overline{\psi}(T)\rangle]\hat{O}|\overline{\psi}(T)\rangle\\&+\frac{i}{\hbar}\,\eta\hat{O}|\overline{\delta\psi}(T)\rangle.\end{aligned} \tag{2.14b}$$

Since we consider $\delta\mathcal{E}(t)$ as an arbitrary variation we can write

$$|\overline{\delta\psi}(t)\rangle\equiv\int_0^T dt\; S_\psi(t,\tau)\,\delta\mathcal{E}(\tau), \tag{2.15a}$$

$$|\overline{\delta\lambda}(t)\rangle\equiv\int_0^T dt\; S_\lambda(t,\tau)\,\delta\mathcal{E}(\tau), \tag{2.15b}$$

where $S_\psi(t,\tau)=|\overline{\delta\psi}(t)\rangle/\delta\mathcal{E}(\tau)$ and $S_\lambda(t,\tau)=|\overline{\delta\lambda}(t)\rangle/\delta\mathcal{E}(\tau)$ are functional derivatives with respect to the field. Insertion of these definitions into Eqs. (2.13a) and (2.13b) and (2.14a) and (2.14b) produces the necessary equations for the determination of these sensitivity coefficients. The existence of these equations and their solutions is sufficient for the analysis here.

If we define the unit operator $\mathcal{I}$ as an integral operator over the Dirac δ function

$$\mathcal{I}f(t)\equiv\int_0^T d\tau\;\delta(t-\tau)f(\tau), \tag{2.16}$$

where $f(t)$ is an arbitrary integrable function over the domain $t\in[0,T]$, then we can reexpress the first part of the right-hand side of Eq. (2.11) as

$$\begin{aligned}(\overline{\delta^2\mathcal{J}})_1\equiv&\int_0^T dt\; W_{\mathcal{E}}(t)\,\delta\mathcal{E}(t)^2\\=&\int_0^T dt\; W_{\mathcal{E}}(t)^{1/2}\delta\mathcal{E}(t)\mathcal{I}W_{\mathcal{E}}(t)^{1/2}\delta\mathcal{E}(t).\end{aligned} \tag{2.17}$$

Separating the simple integral in Eq. (2.17) in this way will be useful below. The rightmost term of Eq. (2.11) can be rewritten by using Eqs. (2.15a) and (2.15b) as follows:

$$\begin{aligned}(\overline{\delta^2\mathcal{J}})_2\equiv&2\int_0^T dt\;\;\delta\mathcal{E}(t)\mathrm{Re}[\langle\overline{\delta\lambda}(t)|\mathcal{D}|\overline{\psi}(t)\rangle\\&+\langle\overline{\lambda}(t)|\mathcal{D}|\overline{\delta\psi}(t)\rangle]\\=&2\int_0^T dt\int_0^T d\tau\;\delta\mathcal{E}(t)\mathrm{Re}[\langle S_\lambda(t,\tau)|\mathcal{D}|\overline{\psi}(t)\rangle\\&+\langle\overline{\lambda}(t)|\mathcal{D}|S_\psi(t,\tau)\rangle]\,\delta\mathcal{E}(\tau),\end{aligned} \tag{2.18}$$

where t and τ are dummy integration variables. Hence, the value of the double integral above remains unchanged when t and τ are interchanged. By using this fact we can write

$$(\overline{\delta^2\mathcal{J}})_2=\int_0^T dt\; W_{\mathcal{E}}(t)^{1/2}\delta\mathcal{E}(t)\mathcal{S}W_{\mathcal{E}}(t)^{1/2}\delta\mathcal{E}(t). \tag{2.19}$$

The stability operator $\mathcal{S}$ is defined over an arbitrary integrable function $f(t)$ as

$$\mathcal{S}f(t) \equiv \int_0^T d\tau\, K(t,\tau) f(\tau), \tag{2.20}$$

where the kernel of the stability operator $\mathcal{K}(t,\tau)$ is explicitly given below as

$$\mathcal{K}(t,\tau) \equiv W_{\mathcal{E}}(t)^{-1/2} W_{\mathcal{E}}(\tau)^{-1/2} \mathrm{Re}[\langle S_\lambda(t,\tau)|\mathcal{D}|\overline{\psi}(t)\rangle + \langle S_\lambda(\tau,t)|\mathcal{D}|\overline{\psi}(\tau)\rangle + \langle \overline{\lambda}(t)|\mathcal{D}|S_\psi(t,\tau)\rangle + \langle \overline{\lambda}(\tau)|\mathcal{D}|S_\psi(\tau,t)\rangle]. \tag{2.21}$$

This kernel function can be interpreted as the t, τ symmetrized value of the functional derivative $\delta\mathcal{E}(t)/\delta\mathcal{E}(\tau)|_c$, for $0<t$, $\tau<1$ where the index c denotes the constraint that the field follow the controlled dynamics. Since control imposes specific structural constraints on the field, this functional derivative deviates from $\delta(t-\tau)$, which is its explicit value when the field is considered as completely arbitrary. This conclusion comes from the fact that Eq. (2.10) is valid only under the condition of system control.

Finally we can write the following equation for the second variation of the cost functional:

$$\overline{\delta^2 \mathcal{J}} = \int_0^T dt W_{\mathcal{E}}(t)^{1/2} \delta\mathcal{E}(t)[\mathcal{I}-\mathcal{S}] W_{\mathcal{E}}(t)^{1/2} \delta\mathcal{E}(t). \tag{2.22}$$

As long as the integral in Eq. (2.22) remains positive, the corresponding optimal solution is a local minimum in the cost functional. Smaller values of $\overline{\delta^2 \mathcal{J}}$ correspond to more robust solutions. The eigenvalues of the stability operator $\mathcal{S}$ determine the optimality and robustness. The spectrum of $\mathcal{S}$ lies on the real axis, as the kernel $\mathcal{K}(t,\tau)$ is real symmetric.

If the stability operator $\mathcal{S}$ has its largest eigenvalue less than 1, then a local minimum for the cost functional is guaranteed at the optimal solution value. For any particular optimal solution, as this largest eigenvalue gets closer to 1, the robustness of the solution increases. The corresponding eigenvectors of $\mathcal{S}$ dictate the temporal variations of the control field $\delta\mathcal{E}(t)$ that produce an associated response for the cost functional $\mathcal{J}$. If the largest eigenvalue of $\mathcal{S}$ exceeds 1, the solution is no longer locally optimal as a minimum of the cost functional. The full analysis of the spectrum of $\mathcal{S}$ will be system dependent and it calls for an elaborate functional analysis. In the following section we will derive a bound on the second variation of the cost functional that can give useful physical insight into optimality and robustness.

III. A QUALITATIVE ROBUSTNESS AND OPTIMALITY ANALYSIS

Considerable physical insight into robustness and optimality can be obtained from the qualitative analysis of the second-order variational relation in Eq. (2.11). We will carry out this analysis by identifying a lower bound of the right-hand side of the equation.

First consider the weight function for the field $W_{\mathcal{E}}(t)$. It must always be positive, except possibly at a finite number of points where it may vanish. This behavior enables us to define the following minimum average value for the weight $W_{\mathcal{E}}(t)$:

$$\overline{W}_{\mathcal{E}} = \min_{f(t)} \left(\frac{\int_0^T dt\; W_{\mathcal{E}}(t) f^2(t)}{\int_0^T dt\; f^2(t)} \right), \tag{3.1}$$

and therefore we have the bound on the first term of Eq. (2.11) as $\int_0^T dt\;\; \delta\mathcal{E}^2(t) W_{\mathcal{E}}(t) \geqslant \overline{W}_{\mathcal{E}} \int_0^T dt\;\; \delta^2\mathcal{E}(t)$. If we use this definition and follow a careful norm analysis for the second term in Eq. (2.11), we can write the following lower bound for the overall second variation of the cost functional as

$$\overline{\delta^2 \mathcal{J}} \geqslant \left(\int_0^T dt[\delta\mathcal{E}(t)]^2 \right)^{1/2} \Bigg[\overline{W}_{\mathcal{E}} \left(\int_0^T dt[\delta\mathcal{E}(t)]^2 \right)^{1/2} - \int_0^T dt[\|\delta\overline{\lambda}(t)\|\; \|\mathcal{D}\overline{\psi}(t)\| + \|\delta\overline{\psi}(t)\|\; \|\mathcal{D}\overline{\lambda}(t)\|] \Bigg]. \tag{3.2}$$

Here the norm on a state vector is defined as

$$\|f(t)\| \equiv \langle f|f\rangle^{1/2}. \tag{3.3}$$

We assume that the dipole operator is bounded by a constant M,

$$\|\mathcal{D}\| \leqslant M, \tag{3.4}$$

over the domain sampled by the dynamics, and then we can write

$$\|\mathcal{D}\overline{\psi}(t)\| \leqslant M \tag{3.5}$$

and

$$\|\mathcal{D}\overline{\lambda}(t)\| \leqslant M \|\overline{\lambda}(t)\|. \tag{3.6}$$

A bound can also be constructed for the Lagrange multiplier. To this end, we can project both sides of Eq. (2.9a) and its complex conjugate upon $-i/\hbar\langle\overline{\lambda}(t)|$ and $i/\hbar|\overline{\lambda}(t)\rangle$, respectively, and add the resulting equations to yield

$$\frac{\partial \|\overline{\lambda}(t)\|^2}{\partial t} = \frac{2i}{\hbar}\, W_p(t) \langle \overline{\psi}(t)|\hat{O}'|\overline{\psi}(t)\rangle \mathrm{Im}[\langle \overline{\lambda}(t)|\hat{O}'|\overline{\psi}(t)\rangle], \tag{3.7}$$

where we used the self-adjointness of the operator $\hat{O}'$.

If we integrate both sides of the last equation over time from t to T, then we can write

$$\|\overline{\lambda}(t)\|^2 = \frac{1}{\hbar^2} [\langle \overline{\psi}(T)|\hat{O}|\overline{\psi}(T)\rangle - \widetilde{O}]^2 \langle \overline{\psi}(T)|\hat{O}^2|\overline{\psi}(T)\rangle - \frac{2i}{\hbar} \int_t^T dt\;\; W_p(t) \langle \overline{\psi}(t)|\hat{O}'|\overline{\psi}(t)\rangle \times \mathrm{Im}[\langle \overline{\lambda}(t)|\hat{O}'|\overline{\psi}(t)\rangle]. \tag{3.8}$$

We will assume that the operators $\hat{O}$ and $\hat{O}'$ are bounded for simplification of the subsequent analysis. Their bounds are given by

$$B_U = \max_{t\in[0,T],f} \langle f(t)|\hat{O}|f(t)\rangle, \quad B_L = \min_{t\in[0,T],f} \langle f(t)|\hat{O}|f(t)\rangle, \tag{3.9a}$$

$$B' = \max_{t\in[0,T],f} \langle f(t)|\hat{O}'|f(t)\rangle, \tag{3.9b}$$

where $\langle f(t)|$ and $|f(t)\rangle$ stand for an arbitrary state vector of unit norm. These definitions make it possible to write

$$[\langle \overline{\psi}(T)|\hat{O}|\overline{\psi}(T)\rangle - \widetilde{O}]^2 \leqslant B_O = \max\{(B_U-\widetilde{O})^2,(B_L-\widetilde{O})^2\}. \tag{3.9c}$$

We further assume that the operator $\hat{O}$ is positive definite, which permits writing the following inequality after some intermediate steps:

$$\|\overline{\lambda}(t)\| \leqslant \frac{B'^2}{\hbar}\mathcal{I}_p + \left[\frac{B_U^2B_O^2}{\hbar^2} + \frac{B'^4}{\hbar^2}\mathcal{I}_p^2\right]^{1/2}, \tag{3.10}$$

where

$$\mathcal{I}_p \equiv \int_0^T dt\ W_p(t) \tag{3.11}$$

A similar treatment of Eq. (2.12a) gives the equation,

$$\frac{\partial\|\delta\psi(t)\|^2}{\partial t} = \frac{2i}{\hbar}\ \delta\mathcal{E}(t)\mathrm{Im}[\langle\delta\psi(t)|\mathcal{D}|\overline{\psi}(t)\rangle]. \tag{3.12}$$

Integration of both sides of this equation over time from 0 to t enables us to conclude that

$$\|\delta\psi(t)\| \leqslant \frac{2}{\hbar}M\left(\int_0^T dt[\delta\mathcal{E}(t)]^2\right)^{1/2}. \tag{3.13}$$

Similar steps can also be taken to conclude the following bound for the first variation of the Lagrange multiplier:

$$\|\delta\lambda(t)\| \leqslant \frac{\mathcal{A}M}{\hbar}\left(\int_0^T dt[\delta\mathcal{E}(t)]^2\right)^{1/2}, \tag{3.14}$$

where

$$\mathcal{A} \equiv \mathcal{A}_1 + \left[\mathcal{A}_1^2 + \frac{4B_U^2[4B_U^2+B_O^2]}{\hbar^2}\right]^{1/2}, \tag{3.15}$$

and

$$\mathcal{A}_1 \equiv \frac{7B'^2}{\hbar}\mathcal{I}_p + \left[\frac{B_U^2B_O^2}{\hbar^2} + \frac{B'^4}{\hbar^2}\mathcal{I}_p^2\right]^{1/2}. \tag{3.16}$$

Exploitation of these relations in Eq. (2.17) yields the following lower bound for the second variation of the cost functional:

$$\overline{\delta^2\mathcal{J}} \geqslant (1-E_r)\overline{W_{\mathcal{E}}}\left(\int_0^T dt[\delta\mathcal{E}(t)]^2\right), \tag{3.17}$$

where

$$E_r \equiv \frac{2TM^2}{\hbar^2\overline{W_{\mathcal{E}}}}\left(\frac{\hbar\mathcal{A}}{2} + B'^2\mathcal{I}_p + [B_U^2B_O^2 + B'^4\mathcal{I}_p^2]^{1/2}\right). \tag{3.18}$$

These equations imply that $E_r<1$ suffices for the existence of a locally optimal control. This result, however, is not a necessary condition. The quality of the bound may be rough, but it does indicate the qualitative relationship of the physical variables dictating optimality and robustness. Within the domain $0<E_r<1$ those values of E_r approaching 1 correspond to more robust solutions as $\overline{\delta^2\mathcal{J}}$ is reduced.

Some interesting qualitative conclusions may be drawn from the structure of Eqs. (3.17) and (3.18). To do so we will assume that $E_r<1$ and the issue of interest is how the physical variables act to increase the robustness by $E_r\rightarrow 1$; an extension of this behavior eventually results in an unacceptable physical solution $E_r>1$ with $\overline{\delta^2\mathcal{J}}<0$, as illustrated in Fig. 1. With these comments in mind we may draw the following conclusions from Eqs. (3.17) and (3.18).

(i) *Objective operator*. Increasing values of B_O, which depends on the objective operator, will enhance the robustness. However, as the control objective is approached, $B_U \approx \widetilde{O}$, a decrease in robustness may be encountered, with the control being accordingly more sensitive to the field variations.

(ii) *Penalty operator*. Increasing the contribution of $\mathcal{B}'^2\mathcal{I}_p$ from the penalty operator will enhance the robustness.

(iii) *Dipole moment operator*. Enhanced robustness occurs with increasing magnitude of the dipole moment operator, apparently, arising due to more effective control regardless of the field strength.

(iv) *Fluence weight*. A decrease of the fluence weight corresponds to an enhancement of robustness. This behavior is associated with a corresponding increase of the field and, hence, stronger control in this regime.

(v) *Control time interval*. Increasing the control time interval enhances robustness. This situation corresponds to the weak-field regime in contrast to point (iv) above.

These observations are physically reasonable, and the behavior in item (ii) is perhaps the most interesting. Enhanced robustness through the introduction of a penalty operator is analogous to the presence of viscous drag acting to stabilize motion of an object moving through a fluid (i.e., a certain degree of drag in the system can be helpful at times). Once again, all of the circumstances in points (i)–(v) when taken beyond a critical limit lead to a nonoptimal control solution. Additional subtleties might arise from the full analysis of the eigenvalues of the stability operator $\mathcal{S}$ rather than the bounding behavior examined here.

IV. CONCLUDING REMARKS

This paper presented a general framework for analyzing the optimality and robustness of any particular quantum control solution. It is shown that both of these issues are dictated by the eigenvalue spectrum of the stability operator $\mathcal{S}$ whose kernel $\mathcal{K}(t,\tau)$ is related to the dynamically constrained functional derivative $\delta\mathcal{E}(t)/\delta\mathcal{E}(\tau)|_c$ for $0<t,\ \tau<T$. No attempt was made to conduct a full functional analysis of this problem in the present paper. Here a bound on the spectrum led to an interesting set of qualitative conditions regarding robustness and optimality. These conditions may serve to qualitatively guide future robust design efforts for the control of quantum systems. In general, the functional analog of the

situation in Fig. 1 may arise with the degree of robustness and the quality of the achieved control in competition with each other. In such a case a judgment would need to be made regarding which factor is more important for the particular system.

ACKNOWLEDGMENTS

The authors acknowledge support from the U.S. Army Research Office, the U.S. Air Force Office of Scientific Research, and the National Science Foundation.

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