Optimal control of quantum systems: Origins of inherent robustness to control field fluctuations

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The impact of control field fluctuations on the optimal manipulation of quantum dynamics phenomena is investigated. The quantum system is driven by an optimal control field, with the physical focus on the evolving expectation value of an observable operator. A relationship is shown to exist between the system dynamics and the control field fluctuations, wherein the process of seeking optimal performance assures an inherent degree of system robustness to such fluctuations. The presence of significant field fluctuations breaks down the evolution of the observable expectation value into a sequence of partially coherent robust steps. Robustness occurs because the optimization process reduces sensitivity to noise-driven quantum system fluctuations by taking advantage of the observable expectation value being bilinear in the evolution operator and its adjoint. The consequences of this inherent robustness are discussed in the light of recent experiments and numerical simulations on the optimal control of quantum phenomena. The analysis in this paper bodes well for the future success of closed-loop quantum optimal control experiments, even in the presence of reasonable levels of field fluctuations.

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I. INTRODUCTION

Over the past decade, a variety of optimal control calculations have been performed for the purposes of exploring the manipulation of quantum phenomena [1,2]. Recently, a number of successful closed-loop optimal control experiments [3–14] have also been carried out where the optimal fields were directly identified in the laboratory using suitable learning control techniques [15–17]. Seeking optimality is natural, as achieving the best possible quantum system performance is always desirable. Of special interest is the contribution of dynamical coherences, as manifested through the manipulation of constructive and destructive quantum wave interferences. There is only an incomplete understanding of the role of such interferences, especially in the most interesting cases involving strong field control [4,7,14], where quantum systems exhibit highly nonlinear field effects. An early point of speculation was that even modest field noise would effectively kill the successful achievement of quantum control in the strong-field regime, where the quantum system would act to amplify the field noise. The intriguing recent experiments operating in this regime give very encouraging evidence that this speculation was incorrect. However, the detailed mechanism for the surprising degree of robustness has remained unclear, especially with successful optimal control being observed in diverse systems [3–14], suggesting that this behavior may be generic.

Several theoretical and experimental studies provide the relevant background for the analysis in this paper. An optimally designed field for the dissociation of hydrogen fluoride showed excellent robustness to field fluctuations [18], and learning control simulations indicate that closed-loop experiments should naturally gravitate towards control fields that produce robustness with respect to the presence of field fluctuations [19,20]. Simulations suggest that explicitly seeking robustness [19] as an additional control criteria can further enhance this stable behavior, and in favorable cases, even with little deterioration in the quality of the attained objective. Most significant are the recent laboratory demonstrations of highly nonlinear intense-field-controlled dissociation and rearrangement of molecules [4,7,14], as well as the manipulation of high harmonic generation [5]. Lurking behind all of these observations is the unknown degree of ''quantum character'' retained in the control process, especially in the presence of field fluctuations. In some special instances, field fluctuations may be helpful [21], but the general expectation is that field noise will be deleterious at least to some extent. The presence of field noise may also influence the rate of convergence or other algorithmic aspects of the laboratory learning control process [17,20].

This paper will explore the relationship between (a) the nature of coherent quantum dynamics, (b) the influence of field fluctuations, (c) the degree of robustness, and (d) the attainment of optimality. It will be shown that the bilinear dependence of quantum expectation values upon the evolution operator and its adjoint has a special serendipitous role in relating all of these points. Detailed simulations of the phenomena involved are difficult to perform, especially for the most interesting complex chemical/physical systems. However, under certain simple assumptions and dynamical considerations, it will be shown that a clear physical picture emerges. The physical picture and its consequences are consistent with current observations and should ultimately aid in providing mechanistic insights into the control of quantum phenomena. Under the most severe degrees of control field fluctuations, the quality of the achieved objective should eventually diminish significantly. However, before that occurs, it will be argued that even the loss of multiple pathway interferences may still lead to successful quantum control processes.

Section II will present an analysis of system behavior when seeking quantum optimal control in the presence of field fluctuations. Section III contains an illustration of the concepts involved. Section IV will discuss the consequences of this behavior for control. Some brief concluding remarks will be given in Sec. V, regarding future studies to amplify on the findings of this paper.

II. OPTIMALLY CONTROLLED QUANTUM DYNAMIC BEHAVIOR

Consider a quantum system with the Hamiltonian $H_0 = H_0 - \mu\varepsilon(t)$, where H_0 is the field-free Hamiltonian, μ is the electric dipole moment, and $\varepsilon(t)$ is the control field. The analysis below also encompasses more general circumstances than just electric dipole interactions, with the only criteria being the existence of a term in the Hamiltonian permitting significant coupling with the control field; for our purposes here, the Hamiltonian above provides a convenient picture to explore the behavior of a quantum system under control. No assumption needs to be made about complete controllability [22] of the quantum system, as it is only sufficient that the target goal be acceptably achieved by some suitable control field. The system initially is described by the density matrix $\rho(0)$, corresponding to a pure or mixed state, and the dynamics is given by

$$i\hbar \frac{\partial \rho}{\partial t} = [H,\rho]. \tag{1}$$

In the present analysis, the system evolution occurs free of any uncontrolled environmental interactions (e.g., collisions). The formal solution to this equation is $\rho(t) = U(t,0)\rho(0)U^{\dagger}(t,0)$, where the time evolution operator $U(t,0)$ satisfies $i\hbar(\partial/\partial t)U = HU$, $U(0,0)=1$. Quantum control is usually specified by prescribing an objective operator O (taken as time-independent here), whose expectation value

$$\langle O(T)\rangle = \mathrm{tr}[\rho(0)U^{\dagger}(T,0)OU(T,0)] \tag{2}$$

at the target time T is the subject for controlled manipulation. The target time may be either a fixed point or $T\to\infty$. Commonly, O is a projection operator into some desired state, although other choices can arise. Regardless of the particular physical objective, it is always natural to pose the goal as one of achieving the best (i.e., the optimal) result for $\langle O(T)\rangle$. This perspective is fundamental to quantum optimal control theory [23], whether carried out as a design process on the computer [1,2] or executed through closed-loop learning techniques directly in the laboratory [3–17,19,20]. Here optimization may refer to maximization, minimization, or some other specified criteria, which is simply summarized as

$$\underset{\varepsilon(t)}{\mathrm{Opt}}\langle O(T)\rangle, \tag{3}$$

where the field is treated as a control function for variation until the optimal objective criteria is met, as best as possible. This process is understood to produce at least one optimal field $\varepsilon_{\mathrm{opt}}(t)$. Any single laboratory experiment would operate with an electric field $\varepsilon(t) = \varepsilon_{\mathrm{opt}}(t) + \delta\varepsilon(t)$, where $\delta\varepsilon(t)$ is a random disturbance around the nominal optimal field $\varepsilon_{\mathrm{opt}}(t)$; in practice, an ensemble of experiments would be performed, collectively associated with the statistics of the ensemble of noise trajectories $\{\delta\varepsilon(t)\}$. Ideally, the experiments would be performed such that $\delta\varepsilon(t)$ satisfies $\|\delta\varepsilon\| \ll \|\varepsilon_{\mathrm{opt}}\|$ under some suitable norm $\|\cdot\|$, in order to reduce the influence of $\delta\varepsilon(t)$ on the control objective $\langle O(T)\rangle$. The significance of the field fluctuations can depend on the overall magnitude of $\varepsilon_{\mathrm{opt}}(t)$ and the degree of nonlinearity of the control process with respect to the field. Small amplitude field fluctuations may have a considerable impact in some cases. On the other hand, experience with optimal control calculations [18–20] and experiments [3–14] shows that there is significant robustness even to the presence of rather large amplitude field fluctuations [18–20] (e.g., $\sim$25% in some simulations). Field fluctuations may occur due to a variety of factors, and in the present work, we will simply assume that $\delta\varepsilon(t)$ is a random variable characterized by a distribution function $P(\delta\varepsilon(t))$, such that

$$\int D[\delta\varepsilon(t)]P(\delta\varepsilon(t)) = 1. \tag{4}$$

This functional integral is over all possible realizations of the field noise trajectories.

In order to explore the influence of the field fluctuations $\{\delta\varepsilon(t)\}$ upon the behavior of the target objective in Eqs. (2) and (3), Fig. 1 depicts the mapping in Eq. (2) from the initial density operator $\rho(0)$ to the final objective operator expectation value $\langle O(T)\rangle$. The initial density operator $\rho(0)$ is transformed to the final objective operator $\langle O(T)\rangle$ through the simultaneous action of $U(T,0)$ and $U^{\dagger}(T,0)$. Either path in the figure may be thought of as an evolution through a sequence of states $|\ell_i\rangle$, $i=1,2,...,n$. These states will be identified in Sec. III as those associated with high quantum evolution phase sensitivity to control field fluctuations; the states will generally not be the traditional intermediate states associated with the various terms of a perturbation expansion for U. The circles along each path in Fig. 1 denote intermediate ''stopping-off'' points, labeled by these states, along the way to the objective; the meaning of this picture will become clear upon the consideration of the influence of field noise $\{\delta\varepsilon(t)\}$. We may rigorously decompose $U(T(0))$ in accord with Fig. 1, as follows:

$$U(T,0) = \sum\!\!\!\!\!\!\int_{\ell_i',\, i=1,\dots,n} U(T,t_n)|\ell_n'\rangle\langle\ell_n'|U(t_n,t_{n-1})|\ell_{n-1}'\rangle \times\langle\ell_{n-1}'|\cdots|\ell_2'\rangle\langle\ell_2'|U(t_2,t_1)|\ell_1'\rangle\langle\ell_1'|U(t_1,0). \tag{5}$$

The symbol $\sum\!\!\!\!\!\!\int$ denotes a summation or integration over the intermediate-state indices, as appropriate. The number of intermediate steps n along the path and the sequence of intermediate times $t_1,t_2,\dots,t_n$ is, in principle, arbitrary, and further discussion on this point is given in Sec. IV C. On physical grounds, it is suggestive to think in terms of a sequence of evolving events under the influence of the electric field $\varepsilon(t)$, $0\leqslant t\leqslant T$, broken into subintervals $\{\varepsilon_0(t), 0\leqslant t \leqslant t_1\}$, $\{\varepsilon_1(t), t_1\leqslant t\leqslant t_2\},\dots,\{\varepsilon_n(t), t_n\leqslant t\leqslant T\}$, such that the full field is a continuous concatenation of the individual pieces $\varepsilon(t) = [\varepsilon_0(t),\varepsilon_1(t),\dots,\varepsilon_n(t)]$ taken in sequence. Each segment of the field carries on a relay process from one intermediate state to another over an associated time interval.

An expression for $U^{\dagger}(T,0)$ analogous to $U(T,0)$ in Eq. (5) may be written, and their combination utilized to represent the structure in Fig. 1:

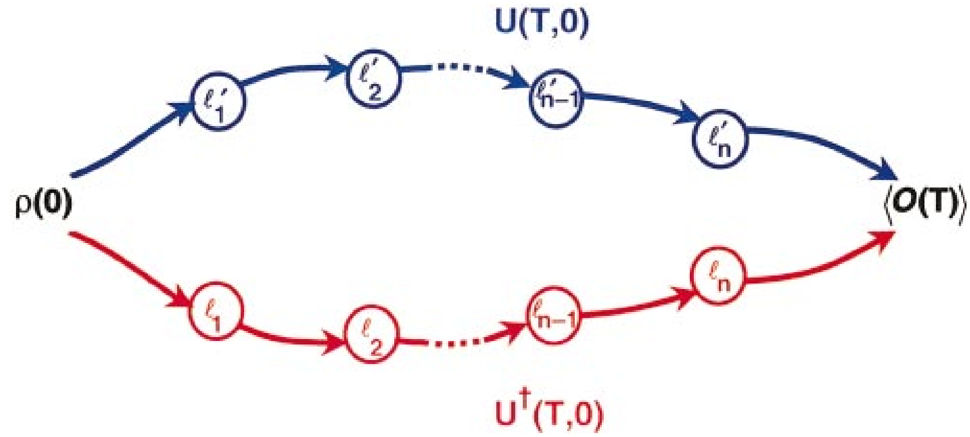

FIG. 1. (Color) A schema depicting the transformation from the initial density operator $\rho(0)$ to the final expectation value $\langle O(T)\rangle$ through a series of dynamical states $|\ell_i\rangle$ and $|\ell_i'\rangle$, $i=1,\ldots,n$ acting as stopping-off points on the excursion. The nature of these states, their total number n, and their location along the excursion are dictated by the ensemble of noise trajectories $\{\delta\varepsilon(t)\}$ associated with the control field. Each stopping-off point acts to break the coherence in the evolution $\rho(0)\rightarrow\langle O(T)\rangle$. The bilinear nature of $\langle O(T)\rangle$ in terms of U and $U^\dagger$ is the origin of the inherent robustness in the evolution, expressed as a set of intermediate coherent steps towards the target $\langle O(T)\rangle$, rather than a total loss of control due to field noise.

$$\mathop{\sum\!\!\!\!\!\!\int}_{\ell_i,i=1,\ldots,n}\ \mathop{\sum\!\!\!\!\!\!\int}_{\ell_i',i=1,\ldots,n}\langle O(T)\rangle=\leftarrow\rho(0)|U^\dagger(t_1,0)|\ell_1\rangle\langle\ell_1|U^\dagger(t_2,t_1)|\ell_2\rangle\langle\ell_2|\cdot\cdots\cdot|\ell_{n-1}\rangle\langle\ell_{n-1}|U^\dagger(t_n,t_{n-1})|\ell_n\rangle$$
$$\times\langle\ell_n|U^\dagger(T,t_n)|O|U(T,t_n)|\ell_n'\rangle\langle\ell_n'|U(t_n,t_{n-1})|\ell_{n-1}'\rangle\langle\ell_{n-1}'|\cdot\cdots\cdots\cdots\cdot|\ell_2'\rangle$$
$$\times\langle\ell_2'|U(t_2,t_1)|\ell_1'\rangle\langle\ell_1'|U(t_1,0)|\rightarrow. \tag{6}$$

Here, the arrows at the beginning and end of the expression imply that these operators are linked together, such that the sequence of matrix elements in Eq. (6) forms a closed loop corresponding to the structure in Fig. 1. Using the definition of Hermitian conjugation along with the modulus and phase decomposition,

$$\langle\ell_q'|U(t_q,t_{q-1})|\ell_{q-1}'\rangle=|\langle\ell_q'|U(t_q,t_{q-1})|\ell_{q-1}'\rangle|\exp[i\phi(\ell_q',\ell_{q-1}')], \tag{7}$$

permits rewriting Eq. (6) in the following fashion:

$$\langle O(T)\rangle=\mathop{\sum\!\!\!\!\!\!\int}_{\ell_i,i=1,\ldots,n}\ \mathop{\sum\!\!\!\!\!\!\int}_{\ell_i',i=1,\ldots,n}\langle\ell_1'|U(t_1,0)|\rho(0)|U^\dagger(t_1,0)|\ell_1\rangle|\langle\ell_2|U(t_2,t_1)|\ell_1\rangle||\langle\ell_2'|U(t_2,t_1)|\ell_1'\rangle|\exp\{i[\phi(\ell_2',\ell_1')$$
$$\vdots$$
$$-\phi(\ell_2,\ell_1)]\}|\langle\ell_n|U(t_n,t_{n-1})|\ell_{n-1}\rangle||\langle\ell_n'|U(t_n,t_{n-1})|\ell_{n-1}'\rangle|\exp\{i[\phi(\ell_n',\ell_{n-1}')-\phi(\ell_n,\ell_{n-1})]\}$$
$$\times\langle\ell_n|U^\dagger(T,t_n)|0|U(T,t_n)|\ell_n'\rangle, \tag{8}$$

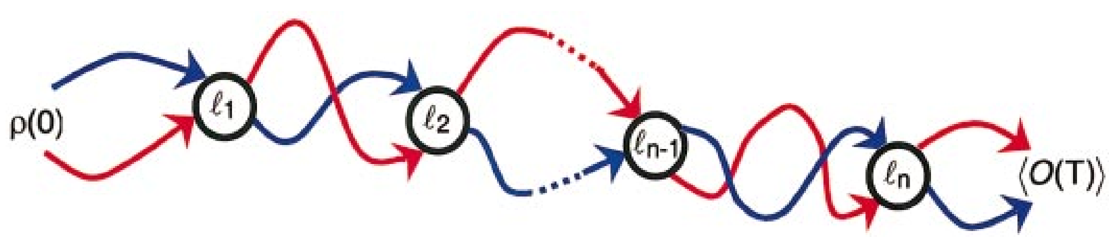

FIG. 2. (Color) The structure arising along the path $\rho(0)\rightarrow\langle O(T)\rangle_{\{\delta\varepsilon\}}$ due to seeking optimal quantum system performance in the presence of control field noise. In spite of noise being present, the optimization process seeks to retain a maximum degree of control through manipulation of constructive/destructive interferences. The result is a reduction of the structure in Fig. 1 down to a sequence of steps shown here and is explicitly expressed in Eq. (15). Within each step (e.g., $\ell_1\rightarrow\ell_2$), full quantum evolutionary coherence is retained while the process is broken in going from one step to the next (e.g., $\ell_1\rightarrow\ell_2$ and then $\ell_2\rightarrow\ell_3$). The interference retained within a step is depicted by the complex interleaving paths.

An arbitrary term in Eq. (8),

$$S_{q,q-1}=|\langle \ell_q|U(t_q,t_{q-1})|\ell_{q-1}\rangle||\langle \ell'_q|U(t_q,t_{q-1})|\ell'_{q-1}\rangle| \times\exp\{i[\phi(\ell'_q,\ell'_{q-1})-\phi(\ell_q,\ell_{q-1})]\}, \tag{9}$$

is a functional of the electric field $\varepsilon_{q-1}(t)$, $t_{q-1}\leqslant t\leqslant t_q$. In Eq. (8), it is understood that each phase $\phi(\ell_p,\ell_{p-1})$, $p=2,...,n$ will generally be a distinct functional of the electric field $\varepsilon_{p-1}(t)$.

In the laboratory, the left-hand side of Eq. (8) would be observed (evaluated) through signal averaging over an ensemble of electric fields centered around the current nominal value $\varepsilon(t)$, with each field realization in the ensemble having a particular noise trajectory $\delta\varepsilon(t)$. In the search for the final optimal field $\varepsilon_{\rm opt}(t)$, a sequence of learning control experiments [15–17,19,20] would be carried out with signal averaging over an ensemble of fields during each cycle of the learning process. Thus, at the end of this laboratory learning control exercise, the final result is an ensemble average of Eq. (8) over the probability distribution function in Eq. (4),

$$\langle 0(T)\rangle_{\{\delta\varepsilon\}}\equiv\int D[\delta\varepsilon(t)]P(\delta\varepsilon(t))\langle O(T)\rangle. \tag{10}$$

The terms in Eq. (8) correspond to the field intervals $\varepsilon(t)=[\varepsilon_0(t),\varepsilon_1(t),\ldots,\varepsilon_n(t)]$, and we will make the assumption that the field fluctuations from one interval to the next are statistically independent of each other, such that

$$P(\delta\varepsilon(t))=\prod_{q=0}^{n}P_q(\delta\varepsilon_q(t)). \tag{11}$$

This factorized form is equivalent to the noise fluctuations having a memory shorter than the time intervals t_q-t_{q-1} for each of the physical evolution steps (i.e., the windows of time between stopping-off points). The presence of correlated noise over extended time periods would change this assumption, and a careful analysis of laser pulse noise will be needed to assess whether a significant modification of the assumption in Eq. (11) is necessary for a more elaborate analysis. Proceeding with the present assumption, combining Eqs. (8), (10), and (11) shows that each of the terms is a separate average over an ensemble of noise trajectories. A typical term in Eq. (9) becomes an average:

$$\langle S_{q,q-1}\rangle_{\{\delta\varepsilon_{q-1}\}}=\int D[\delta\varepsilon_{q-1}(t)]P_{q-1}(\delta\varepsilon_{q-1}(t)) \times S_{q,q-1}([\delta\varepsilon_{q-1}(t)]), \tag{12}$$

where the explicit functional dependence of $S_{q,q-1}$ in the integrand upon the field fluctuations is indicated by $[\delta\varepsilon_{q-1}(t)]$. We will make the reasonable assumption that the most sensitive functional dependence on the field fluctuations in Eq. (9) arises from the phase factors, rather than the moduli. To start the averaging process, consider first the term involving $\rho(0)$ rewritten in the following form:

$$S_1=|\langle \ell'_1|U(t_1,0)|\rho(0)|U^\dagger(t_1,0)|\ell_1\rangle| \times\exp\{i[\phi(\ell'_1)-\phi(\ell_1)]\}. \tag{13}$$

The particular phase structure in Eq. (13) may be argued to exist in the physically relevant extreme cases of $\rho(0)$ being either a pure state $|\psi(0)\rangle\langle\psi(0)|$ or an incoherent mixed state $\Sigma_j|j\rangle\rho_{jj}(0)\langle j|$, where $\rho_{jj}(0)$ is the initial population in the jth state. The phase difference in Eq. (13) can be a sensitive function of $(\ell'_1-\ell_1)$ when there are significant field fluctuations, and it is expected under these conditions that the region $\ell'_1\simeq\ell_1$ will be most important. Expanding the ℓ'_1 dependence of the phase factor about ℓ_1 and keeping only the lowest-order term produces the phase factor $\phi(\ell'_1)-\phi(\ell_1)\simeq\phi'(\ell_1)(\ell'_1-\ell_1)$, where the prime on the phase denotes differentiation. Over a moderate interval for variation of $[\delta\varepsilon_0(t)]$, we may consider the differential probability relation as uniform in the phase derivative, $D[\delta\varepsilon_0(t)]P_0(\delta\varepsilon_0(t))\approx(1/2\pi)d\phi'(\ell_1)$. Thus, combining all of these statements and taking the modulus in Eq. (13) as slowly varying with respect to field fluctuations leads to the result

$$\langle S_1\rangle_{\{\delta\varepsilon_0\}}\simeq|\langle \ell_1|U(t_1,0)|\rho(0)|U^\dagger(t_1,0)|\ell_1\rangle|\delta(\ell'_1-\ell_1), \tag{14}$$

where $\delta(\cdot)$ is a Dirac delta function. The same arguments may be applied sequentially to all of the terms in Eq. (8), with each of them producing a new Dirac delta function amongst the quantum numbers. When considering the generic term in Eq. (9), the relation $\ell'_{q-1}=\ell_{q-1}$ will have already been established from the ensemble average over the previous term. The overall ensemble averaging process will yield a sequence of delta functions, $\Pi_{q=1}^n\delta(\ell'_q-\ell_q)$. Thus, we may express the average in Eq. (10) over Eq. (8) as having the following final form, assuming that the quantum indices ℓ_q, $q=1,...,n$ take a sufficiently dense set of values to be integrated over,

$$\langle O(T)\rangle_{\{\delta\varepsilon\}}\simeq\sum\!\!\!\!\!\!\int_{\ell_i,i=1,...,n}|\langle \ell_1|U(t_1,0)|\rho(0)|U^\dagger(t_1,0)|\ell_1\rangle| \times|\langle \ell_1U(t_1,t_2)|\ell_2\rangle|^2\cdots|\langle \ell_{n-1}|U(t_{n-1},t_n) \times|\ell_n\rangle|^2\langle \ell_n|U^\dagger(T,t_n)|O|U(T,t_n)|\ell_n\rangle. \tag{15}$$

The structure of the expression in Eq. (15) is shown in Fig. 2. By comparison with Fig. 1, it is evident that the process of seeking optimally controlled system performance in the presence of field noise has broken the evolution $\rho(0)\to\langle O(T)\rangle_{\{\delta\varepsilon\}}$ into a sequence of steps. Coherence is fully maintained within each step (e.g., $|\langle \ell_1|U(t_1,t_2)|\ell_2\rangle|^2$), but is broken in going from one step to the next. The following sections will consider the structure of Eq. (15) and its physical implications for quantum control.

III. AN ILLUSTRATION

The control of any particular quantum system will have its own special nuances, but a very common circumstance is for the control field $\varepsilon(t)$ over the domain $0 \leqslant t \leqslant T$ to have at least three windows $0 \rightarrow t_1$, $t_1 \rightarrow t_2$, and $t_2 \rightarrow T$, where the field is very intense during the intermediate time $t_1 \leqslant t \leqslant t_2$. Additional windows of high intensity may also occur, and the analysis below may be readily extended to such a generalization. For the purposes of illustration, it will be assumed that the window $t_1 \leqslant t \leqslant t_2$ is sufficiently short, such that the dynamics may be taken as sudden during this interval. Furthermore, it is reasonable to take the field fluctuations $\delta\varepsilon(t)$ as especially intense during this window, since a correlation will likely exist between the local-field amplitude and its fluctuation magnitude. The influence of field fluctuations on the evolving dynamics is sensitive to the magnitude of the fluctuations $\|\delta\varepsilon(t)\|$, and accordingly, the illustration will consider the fluctuations as significant only during the intermediate window $t_1 \leqslant t \leqslant t_2$. The system starts out in a pure state $\rho(0) = |i\rangle\langle i|$, with the objective $O = |f\rangle\langle f|$ being a projection operator.

The observable expectation value at the target time becomes

$$O(T) = \langle f|U(T,0)|i\rangle\langle i|U^{\dagger}(T,0)|f\rangle \tag{16}$$

with $U(T,0) = U(T,t_2)U(t_2,t_1)U(t_1,0)$. The time evolution operator $U(T,0)$ is broken into three steps in keeping with the nature of the control field described above. In addition, due to the sudden dynamics over the period $t_1 \leqslant t \leqslant t_2$, with the Hamiltonian $H = H_0 - \mu\varepsilon(t)$, it follows that

$$U(t_2,t_1) = \exp[-i\mu(\overline{\varepsilon} + \overline{\delta\varepsilon})/\hbar], \tag{17}$$

where $\overline{\varepsilon} = \int_{t_1}^{t_2} \varepsilon(t)dt$ and $\overline{\delta\varepsilon} = \int_{t_1}^{t_2} \delta\varepsilon(t)dt$. As $\delta\varepsilon(t)$ is a random variable, we may similarly treat $\overline{\delta\varepsilon}$ as a new random variable for statistical averaging. Combining these statements, the expression in Eqs. (6) and (10) becomes

$$\begin{aligned}\langle 0(T)\rangle_{\{\overline{\delta\varepsilon}\}} = &\sum_{\ell_1\ell_1'}\sum_{\ell_2\ell_2'} \langle f|U(T,t_2)|\ell_2\rangle\langle \ell_2'|U^{\dagger}(T,t_2)|f\rangle \\ &\times\langle \ell_1|U(t_1,0)|i\rangle\langle i|U^{\dagger}(t_1,0)|\ell_1'\rangle \\ &\times\int d[\overline{\delta\varepsilon}]P(\overline{\delta\varepsilon})\langle \ell_2|U(t_2,t_1)|\ell_1\rangle \\ &\times\langle \ell_1'|U^{\dagger}(t_2,t_1|\ell_2'\rangle,\end{aligned} \tag{18}$$

Here, for convenience, the intermediate states $\{|\ell\rangle\}$ are taken as discrete. The averaging operation over the mean-field amplitude fluctuations $\overline{\delta\varepsilon}$ is based on Eq. (7), which reduces to

$$\langle \ell_2|U(t_2,t_1)|\ell_1\rangle = |\langle \ell_2|U(t_2,t_1)|\ell_1\rangle|\exp[i\phi(\ell_2,\ell_1)]. \tag{19}$$

The analysis in Eqs. (8)–(14) is most reliable when the modulus is slowly varying over $\{\overline{\delta\varepsilon}\}$, while the phase has the dominant variation. This behavior can be examined in detail for the model in this illustration. The intermediate states $\{|\ell\rangle\}$ are assumed to be complete but otherwise arbitrary. However, given the structure of Eq. (17), it is natural to consider $\{|\ell\rangle\}$ as eigenstates of the dipole operator

$$\mu|\ell\rangle = \lambda_\ell|\ell\rangle. \tag{20}$$

For now, it will be assumed that the spectrum $\{\lambda_\ell\}$ is nondegenerate; the relaxation of this assumption will be permitted later. Thus, Eq. (19) reduces to

$$\langle \ell_2|U(t_2,t_1)|\ell_1\rangle = \delta_{\ell_1\ell_1}\exp[-i\lambda_{\ell_1}(\overline{\varepsilon} + \overline{\delta\varepsilon})/\hbar], \tag{21}$$

and the integral over the field fluctuations in Eq. (18) becomes

$$\begin{aligned}\int d[\overline{\delta\varepsilon}]P[\overline{\delta\varepsilon}]\delta_{\ell_1\ell_2}\delta_{\ell_i'\ell_2'}\exp[-i(\lambda_{\ell_1} - \lambda_{\ell_1'})(\overline{\varepsilon} + \overline{\delta\varepsilon})/\hbar] \\ \approx \delta_{\ell_1\ell_2}\delta_{\ell_1'\ell_2'}\delta_{\ell_1\ell_1'}.\end{aligned} \tag{22}$$

In the evaluation of the integral, the field fluctuations are taken as sufficiently large, to satisfy the criteria $|\lambda_{\ell_1} - \lambda_{\ell_1'}|\,\|\overline{\delta\varepsilon}\| \gg \pi$ for any $\ell_1 \neq \ell_1'$, where $\|\overline{\delta\varepsilon}\|$ refers to the norm of $\overline{\delta\varepsilon}$ over the domain of variation allowed by $P[\overline{\delta\varepsilon}]$. Utilizing Eqs. (21) and (22) in Eq. (18) finally produces

$$\langle 0(T)\rangle_{\{\overline{\delta\varepsilon}\}} \approx \sum_{\ell} |\langle f|U(T,t_2)|\ell\rangle|^2|\langle \ell|U(t_1,0)|i\rangle|^2. \tag{23}$$

This result is a special case of Eq. (15) under the specified dynamical conditions.

It is evident from Eq. (23) that the field fluctuations over the short period of intense sudden dynamics result in a small stopping-off window, $t_1 \leqslant t \leqslant t_2$, and a loss of coherent linkage across that window. This total loss of coherent linkage would be relaxed under the conditions that either $\|\overline{\delta\varepsilon}\|$ is reduced in magnitude, or the dipole eigenvalue difference $|\lambda_{\ell_1} - \lambda_{\ell_1'}|$ is sufficiently small for some $\ell_1 \neq \ell_1'$, such that $|\lambda_{\ell_1} - \lambda_{\ell_1'}|\|\overline{\delta\varepsilon}\| \lessapprox \pi$. In the limit that the latter quantity becomes very small (e.g., some of the eigenvalues are small or degenerate, or $\|\overline{\delta\varepsilon}\|$ is small), the full coherence linkage across the window $t_1 \leqslant t \leqslant t_2$ should be resurrected. Note that if the entire domain $0 \leqslant t \leqslant T$ corresponds to a very intense ultrashort sudden control interval with sufficiently intense field fluctuations (and a nondegenerate spectrum $\{\lambda_\ell\}$), then the ensemble-averaged dynamics reduces to the form

$$\langle O(T)\rangle_{\{\overline{\delta\varepsilon}\}} \rightarrow \sum_{\ell} |\langle f|\ell\rangle|^2|\langle \ell|i\rangle|^2, \tag{24}$$

corresponding to a statistical outcome and an effective loss of control. When operating with intense fields, these results suggest the existence of a tradeoff. Dynamical control with intense fields has certain attractive features (e.g., the lifting of constraining resonant conditions), but significant field fluctuations can have a deleterious effect on the control process. Generally, a soft graduated influence of field noise is expected, with the conditions leading to Eq. (15) occurring, at most, at a limited number of points or windows during the controlled evolution. Furthermore, the example in this section shows how a partial flow of coherence can pass through the stopping-off points, depending the details of H_0, μ, $\varepsilon(t)$, and $\|\delta\varepsilon\|$.

IV. DISCUSSION

This section will address a set of interrelated topics to explain the physical consequences of the result in Eq. (15), and connect it to the behavior found in recent laboratory experiments and computer simulations on optimally controlled quantum systems.

A. Exploiting constructive and destructive quantum wave interferences

A basic premise underlying the manipulation of quantum systems is that the best control results will be achieved in the circumstances permitting the maximum use of constructive and destructive (C/D) interferences to discriminate amongst the desired and undesired product channels [24]. The presence of noise $\delta\varepsilon(t)$ should diminish the ability to take full advantage of C/D interferences. The ideal limit is the circumstance of $P(\delta\varepsilon(t))\to\delta[\delta\varepsilon(t)]$, which corresponds to no field noise, with $\delta[\cdot]$ being a Dirac delta function. In this limit, the result found in Eq. (15) will reduce to that of a single term (i.e., there will be no intermediate stop-off points), corresponding to the original expression

$$\langle O(T)\rangle = \mathrm{tr}[\rho(0)U^{\dagger}(T,0)OU(T,0)] \tag{25}$$

without further reduction. The formulation in Eq. (15) strictly applies to the case of there at least being a single intermediate stop-off point, $n\geqslant 1$, on the path $\rho(0)\Rightarrow\langle O\rangle$. In the laboratory, there will always be a finite amount of field noise, thereby likely corresponding to the presence of one or more intermediate stopping-off points on the control pathway in Fig. 2 and Eq. (15). Each of these points break the C/D interference process into subpieces, likely resulting in less than full control. Furthermore, the influence of field fluctuation may lead to a gradual diminution of *C/D* effects, as indicated in sec. III.

B. Seeking optimality and achieving robustness

With the ability to reliably shape control pulses [25] and exploit closed-loop learning algorithms [15–17,19,20], the realization of optimal control in the laboratory is now a demonstrated capability [3–14]. The search for optimality in this process will attempt to drive up the degree of C/D interference manipulation, while assuring that the control results are as robust as possible to field noise. Although some serendipitous circumstances may permit good degrees of robustness simultaneously with extensive exploitation of C/D interferences for high product yields [21], the general expectation is that these two goals will be competitive with each other. The overall structure in Eq. (15) and Fig. 2 arises due to ensemble averaging over a finite level of field noise. Thus, seeking optimal performance leads to noise-induced stop-off points on that path $\rho(0)\Rightarrow\langle O(T)\rangle_{\{\delta\varepsilon\}}$ to assure a degree of robustness by eliminating those quantum phases [c.f., Eq. (7)] which have significant sensitivity to field noise. Some optimal control field design calculations [18] were also shown to have unusual levels of robustness to noise, which may indicate that the associated optimally controlled dynamics was broken into modular units according to Eq. (15).

The stable structure of Eq. (15) suggests that reasonable levels of field noise, even at high field intensities, may not result in a catastrophic loss of control. The well-defined modular path between the initial state and the objective in Eq. (15) is expected to inherently produce a more robust control process at the expense of some measured loss of fidelity from drawing on C/D interferences. Perhaps the best evidence for this behavior is the success of the recent high-field quantum control experiments involving molecular dissociation and rearrangement [4,7,14], as well as selective high harmonic generation [5]. Early speculation suggested that selective quantum control in this regime would be of poor quality due to the quantum system effectively acting as an amplifier of even modest field noise resulting from the highly nonlinear dependence upon the field. However, operating under optimal control, the experimental findings and the stable structure in Eq. (15) and Fig. 2 demonstrate otherwise.

One limiting class of "control" experiments will be those carried out without any benefit of optimal field training to meet the objectives. An example along these lines is high-field multiphoton ionization of atoms and molecules using bandwidth limited pulses [26]. The expectation is that this circumstance will utilize little, if any, beneficial C/D interferences, but perhaps exhibit a maximal degree of robustness for the ionization signal that is generated. This regime should correspond to utilizing a maximum number of intermediate stop-off points in Eq. (15), producing a ladder of stepwise transitions to the ionization continuum. This behavior appears to be operative in nonoptimal multiphoton ionization [26].

C. The stationary stopping-off points on the way to optimal control

Consider now the number of steps n on the way to the target in Eq. (15), as well as their location in time and quantum number space $\{t_q,\ell_q\}$. Based on the phase averaging arguments leading to Eq. (15), coupled with the attainment of optimality drawing on the highest possible degree of C/D interference manipulation, we may identify the sequence of points $\{t_q,\ell_q\}$ as locations where there is *high* quantum evolution phase sensitivity to field fluctuations. In order to optimally achieve the control objective with good robustness, the quantum evolution phase sensitivity is diminished at $\{t_q,\ell_q\}$, by cancellation of the pairs of phases at the analogous points along the evolution of $U(T,0)$ and $U^{\dagger}(T,0)$, as indicated in Figs. 1 and 2 and Eq. (9). In turn, the evolution over the interval $\ell_{q-1}\to\ell_q$ between two such stopping-off points corresponds to a domain of lesser sensitivity to field noise, and hence, an inherent degree of robustness, thereby permitting some exploitation of C/D interference through $|\langle\ell_q|U(t_q,t_{q-1})|\ell_{q-1}\rangle|^2$ on the way towards the desired objective. Increasing noise levels should lead to more such intermediate phase sensitive points $\{t_q,\ell_q\}$, with the limit ultimately reducing the dynamics to a sequence of incoherently coupled steps (e.g., in the case of dipole coupling, each step in this limit would correspond to a particular matrix element $\mu_{q,q-1}$). Such a chain of simple steps is still quantum mechanical, as governed by the system selection rules.

The physical nature of the intermediate states $|\ell_q\rangle$, $q=1,2,...,n$ is dictated by the optimal control process seeking the best system performance. Thus, these states are the set that is consistent with the physical objectives and the attainable dynamics, so as to produce optimal performance by best eliminating the field fluctuation-driven quantum evolution phase sensitivity. These intermediate states might be members of the eigenstates of H_0 or superpositions of them to form virtual states. The guidance is strictly driven by seeking optimality. Only the existence of the states is necessary to provide a basis to explain the robustness with respect to field noise.

Finally, to aid in understanding the meaning of the intermediate steps, it is useful to consider the special case of $\rho(0)=|i\rangle\langle i|$ and $O=|f\rangle\langle f|$ corresponding to control of population transfer for $|i\rangle\rightarrow|f\rangle$. In this case, Eq. (15) simply reduces to

$$\sum_{\ell_j, j=1,...,n} \prod_{j=1}^{n+1} |\langle \ell_j|U(t_j,t_{j-1})|\ell_{j-1}\rangle|^2, \tag{26}$$

with $|\ell_0\rangle\equiv|i\rangle$; $t_{n+1}\equiv T$, $|\ell_{n+1}\rangle\equiv|f\rangle$. Thus, the final probability of occupying the state $|f\rangle$ is the product of the individual probabilities for making intermediate transitions. Despite some loss of C/D manipulation in Eq. (26), the final population yield could still approach unity if each step along the way from $i\rightarrow f$ is highly efficient. This prospect points out that caution may be called for with regard to physically interpreting successful quantum control experiments in terms of their fully drawing on C/D interferences.

In considering Eq. (26), as well as the more general result in Eq. (15), no further specification of the intermediate times is necessary. Although the discussion here implies that there would likely be a correlation between these times and the associated intermediate stopping-off points, an expression analogous to Eq. (15) may also be derived involving integration over the intermediate times.

D. Evolution in the presence of field fluctuations

After attaining the optimal control field $\varepsilon_{\mathrm{opt}}(t)$, its application to the system under ideal noise-free conditions would produce the time evolution described by the operator $U_{\mathrm{opt}}(t,0)$. In practice, each particular control field $\varepsilon_i(t)=\varepsilon_{\mathrm{opt}}(t)+\delta\varepsilon_i(t)$ from a laboratory ensemble would have noise fluctuations around the optimal field, and would have its own associated unitary evolution operator $U_i(t)$, $i=1,2,...$. By virtue of the optimality and robustness of the dynamics under $\varepsilon_{\mathrm{opt}}(t)$, this ensemble of evolution operators is expected to be reasonably stable with respect to noise, except at the set of stopping-off points over the interval $0\leqslant t\leqslant T$. In the local neighborhood of the stopping-off points, high sensitivity to field noise will occur for each operator $U_i(t)$. However, the expectation value $\langle O(T)\rangle_i$ determined from a particular field $\varepsilon_i(t)$ should have diminished noise sensitivity, and the ensemble average in Eq. (15) should exhibit good robustness to field noise.

V. CONCLUSION

This paper presented a general analysis of optimal quantum system control, with the aim of investigating the overall impact of field noise on the control process. It was argued that a relationship exists between (a) the nature of quantum dynamics being bilinear in U and $U^{\dagger}$, (b) the presence of field fluctuations, (c) the attainment of optimality, and (d) the robustness of the control process. Although noise is expected to generally have a deleterious effect on achieving control, especially in the nonperturbative regime, the analysis showed that good control selectivity may still remain, with the power of optimality fighting to achieve the best results possible. To push this analysis further, it would be very desirable to carefully assess the nature of shaped laser-pulse field noise, and ideally, vary the noise in specific ways to observe its impact on the optimal control process. Furthermore, it would also be valuable to introduce explicit techniques to identify the actual quantum pathways linking the initial and final states [27]. In some limiting cases [e.g., Eq. (26)], it may be possible to construct the observed control event from a sequence of observations in separate subcontrol experiments.

The inherent degree of robustness evident in the optimal control of quantum systems is very encouraging for future applications. In most of these applications, the physical focus is on $\langle O(T)\rangle$, and often, even a modest degree of stable control would be quite acceptable. A notable exception may arise in quantum information science [28], where one focus is on $U(t)$, $t\geqslant 0$ acting as a functional quantum ''machine'' (e.g., a quantum computer). In this case, the presence of dynamical quantum phases sensitive to field noise may be troublesome. But again, seeking optimal performance should also provide the best operational framework.

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