Quantum optimal control theory in the linear response formalism

Alberto Castro*
Institute for Biocomputation and Physics of Complex Systems (BIFI) and Zaragoza Center for Advanced Modelling (ZCAM), University of Zaragoza, ES-50009 Zaragoza, Spain

I. V. Tokatly†
Nano-Bio Spectroscopy Group and ETSF Scientific Development Centre, Departamento de Física de Materiales, Universidad del País Vasco UPV/EHU, ES-20018 San Sebastián, Spain and IKERBASQUE, Basque Foundation for Science, ES-48011 Bilbao, Spain
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Quantum optimal control theory (QOCT) aims at finding an external field that drives a quantum system in such a way that optimally achieves some predefined target. In practice, this normally means optimizing the value of some observable, a so-called merit function. In consequence, a key part of the theory is a set of equations, which provides the gradient of the merit function with respect to parameters that control the shape of the driving field. We show that these equations can be straightforwardly derived using the standard linear response theory, only requiring a minor generalization: the unperturbed Hamiltonian is allowed to be time dependent. As a result, the aforementioned gradients are identified with certain response functions. This identification leads to a natural reformulation of QOCT in terms of the Keldysh contour formalism of the quantum many-body theory. In particular, the gradients of the merit function can be calculated using the diagrammatic technique for nonequilibrium Green's functions, which should be helpful in the application of QOCT to computationally difficult many-electron problems.

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I. INTRODUCTION

Quantum optimal control theory (QOCT) [1,2] is concerned with finding a time-dependent external field that drives a given quantum system to optimally achieve some predefined target that depends on the manner in which the system evolves.[1] For example, a target can be the population of some excited state at the final time of the propagation, but many other options are possible. The theory can be regarded as a branch of the *classical* control theories developed mostly in the fields of mathematics and engineering [3,4]. The quantum discipline was born in the late 1980s [5–7] as the most complete theoretical framework capable of addressing the nascent experimental field of quantum control (or *coherent control*) [8]. The range of applications of quantum control is growing very fast, thanks to the progress in the ultrafast laser-pulse generation and pulse-shaping techniques [9], as well as to the development of adaptive feedback control schemes [10,11]. Typical examples of applications are the control of the population of excited states in molecules [11], optimization of high-harmonic generation [12], optimization of selective photodissociation of molecules [13], optimization of multiphoton ionization of atoms [14], enhancement of electron transfer in dye-sensitized solar cells [15], etc.

At the formal level, the central problem of QOCT is to maximize an expectation value of some operator, usually known as a merit (or target) function, the input of which is the external field that needs to be optimally shaped. The field is normally parametrized either by a discrete set of real-valued "control" parameters, or, in a more general setting, by continuous functions of time. In the latter case, one usually speaks of target functionals. In most cases, the optimization algorithm will require both the computation of the merit function and of its gradient with respect to control parameters. Therefore, an expression and computational strategy for this gradient constitutes one of the most important parts of QOCT.

The usual derivation of expressions for the gradient of the merit function proceeds via the definition of a Lagrangian functional, and of a "Lagrange multiplier" wave function (see, for example, Refs. [6,16]). It leads to an expression for the gradient that involves the forward propagation of the system wave function, and the backward propagation of the new Lagrange multiplier wave function. At this point, it is worth noting that the presence of forward and backward time propagations is a general feature of the quantum kinetic theory, which can be conveniently formulated as a propagation along the Keldysh-Schwinger closed-time contour [17,18]. Therefore, it is natural to expect that there is a connection between QOCT and the Keldysh contour formulation of the quantum dynamics. In this paper, we make this connection explicit by reexamining the derivation of the expression of the gradient (or functional derivative) of the target functional.

Our main simple observation is that the differentiation of a target observable with respect to a control parameter is identical to computing a change of that observable induced by a corresponding perturbation in the Hamiltonian. Thus, the problem of calculating the gradient of the merit function reduces to a generalized form of linear response theory (LRT),

*acastro@bifi.es
†ilyatokatly@ehu.es

[1]Perhaps, more generally, the optimization can also be done with respect to *internal* parameters defining the system itself, and not necessarily defining an external field. However, most applications are for this latter case.

in which the unperturbed Hamiltonian is no longer static but depends on time. The formalism of LRT can then be directly applied, and we straightforwardly recover the very same expressions that one reaches in the "traditional" way. However, these expressions can then be regarded as response functions represented by certain retarded correlation functions. We emphasize that this rederivation is not a mere academic exercise since the new interpretation of the gradient as a response function suggests immediately the use of the known approximations to this object. In particular, by relating the retarded response function to a contour-ordered correlation function, we can apply well-developed methods and approximations of the nonequilibrium many-body perturbation theory to QOCT for many-electron systems [17,19,20].

The latter is an especially important aspect since the treatment of many-electron systems is notoriously difficult; yet, the direct control of electrons is an area of growing interest due to the advances in laser pulses of strong intensity and ultrashort durations, in the attosecond range: the scale of the electronic movements. In order to theoretically study a direct control of electronic motion, it is necessary to have a predictive (*ab initio*) yet computational tractable scheme, in combination with QOCT. Some possibilities have been recently put forward such as (multiconfiguration) time-dependent Hartree Fock [21] and time-dependent density functional theory [22]. Here, we propose a new possibility, based on nonequilibrium many-body Green's function theory.

The structure of the paper is the following. In Sec. II, we derive the gradient QOCT equations in the formalism of LRT. To make this paper self-contained, the slightly generalized basic LRT results needed for this purpose are presented in the Appendix. Section III elaborates on the equations derived in Sec. II by proposing a QOCT scheme for many-body systems, based on the Keldysh contour formalism and on standard approximations in nonequilibrium many-body Green's function theory. We conclude by summarizing our results, discussing directions for further research, and the possibility of numerical implementation of our many-body version of the QOCT.

II. BASIC QOCT EQUATIONS IN THE LINEAR RESPONSE THEORY LANGUAGE

Let us consider a quantum system described by its density matrix $\hat{\rho}(t)$ and governed, in the time interval $[t_0,t_f]$, by a von Neumann equation in the form

$$\frac{\partial}{\partial t}\hat{\rho}(t) = -i[\hat{H}[u](t),\hat{\rho}(t)]\,, \tag{1}$$

$$\hat{\rho}(t_0) = \hat{\rho}_0\,, \tag{2}$$

where the Hamiltonian is given by[2]

$$\hat{H}[u](t) = \hat{\mathcal{H}} + \epsilon[u](t)\hat{V}\,. \tag{3}$$

[2]Equation 3 prescribes a particular form of the Hamiltonian, namely, a linear dependence of it with the control field $\epsilon[u]$. This is perhaps the most common case, and may describe, for example, a laser pulse interacting with an atom or molecule. However, the results that follow do not rely on this particular choice, and could be derived with a generic dependence $\hat{H}[u]$.

The Hamiltonian piece $\hat{\mathcal{H}}$ is static, and $\epsilon[u](t)$ is a time-dependent function the precise form of which is determined by a set of parameters that we will denote, collectively, u. The operator $\hat{V}$ represents the coupling of the system with an external field, e.g., if we think of an atom or molecule irradiated by a laser pulse, the dipole operator. Evidently, a particular choice of the *control* u leads to a system evolution $u \to \hat{\rho}[u](t)$.

We wish to find the values of u that maximize the value of the expectation value of some observable $\hat{A}$ at the end of the propagation. In other words, we want to find the maximum of the function

$$G[u] = \mathrm{Tr}\{\hat{\rho}[u](t_f)\hat{A}\}\,. \tag{4}$$

In order to find the maximum, the best way is to be able to compute the gradient of G. The problem that we face, therefore, is that of finding a suitable expression for this gradient.

Assuming that there is only one parameter u (the generalization to more than one is trivial),

$$\frac{\partial G}{\partial u}[u] = \lim_{\Delta u\to 0}\Delta u^{-1}(G[u+\Delta u]-G[u])\,. \tag{5}$$

Note that $\hat{\rho}[u]$ corresponds to the propagation of the system with the Hamiltonian given in Eq. (3), whereas $\hat{\rho}[u+\Delta u]$ corresponds to the propagation of the system with the Hamiltonian

$$\hat{H}[u+\Delta u](t) = \hat{H}[u](t) + \Delta u\frac{\partial\epsilon}{\partial u}[u](t)\hat{V} \tag{6}$$

to first order in Δu. Now, we can use directly the LRT result introduced in the Appendix by making the identifications

$$\hat{H}_0(t) = \hat{H}[u](t), \quad f(t) = \Delta u\frac{\partial\epsilon}{\partial u}[u](t)\,. \tag{7}$$

Therefore, we just need to apply Eqs. (A12) and (A13) to arrive at

$$\frac{\partial G}{\partial u}[u] = \int_{t_0}^{\infty} d\tau\ \frac{\partial\epsilon}{\partial u}[u](\tau)\chi_{\hat{A},\hat{V}}(t_f,\tau), \tag{8}$$

where

$$\chi_{\hat{A},\hat{V}}(t_f,\tau) = -i\theta(t_f-\tau)\mathrm{Tr}\{\hat{\rho}(t_0)[\hat{A}_H(t_f),\hat{V}_H(\tau)]\} \tag{9}$$

is the response function for the $(\hat{A},\hat{V})$ operators. Inside the commutator, these operators appear in the Heisenberg representation, defined by

$$\hat{O}_H(t) = \hat{U}^{\dagger}(t,t_0)\hat{O}\hat{U}(t,t_0) \tag{10}$$

for any observable $\hat{O}$, and where $\hat{U}(t,t_0)$ is the propagator corresponding to the $\hat{H}[u](t)$ Hamiltonian. Equation (8) clearly manifests how the gradient is nothing else than a response function, albeit a generalized one. It corresponds to the response of a system driven by a time-dependent Hamiltonian to a modification of this Hamiltonian. It remains now to see how this result is equivalent to the expressions obtained in a different manner with the usual QOCT technique. For that purpose, we define an operator

$$\hat{A}[u](\tau) = \hat{U}(\tau,t_f)\hat{A}\hat{U}(t_f,\tau)\,, \tag{11}$$

$$\hat{A}[u](t_f) = \hat{A}\,, \tag{12}$$

which can also be written as the solution to the differential equation

$$\frac{\partial}{\partial t}\hat{A}[u](t) = -i[\hat{H}[u](t), \hat{A}[u](t)]\,, \tag{13}$$

$$\hat{A}[u](t_f) = \hat{A}\,. \tag{14}$$

By using this new auxiliary object, and after a little manipulation of Eq. (8) one arrives at

$$\frac{\partial G}{\partial u}[u] = -i\int_{t_0}^{t_f} d\tau\ \frac{\partial \epsilon}{\partial u}[u](\tau)\mathrm{Tr}\{\hat{\rho}[u](\tau)[\hat{A}[u](\tau), \hat{V}]\}. \tag{15}$$

Equations (13), (14), and (15), together with the original propagation equation for $\hat{\rho}[u](t)$, are the "QOCT equations," usually derived in a different way (through the definition of a Lagrangian function). Algorithmically, the computation of the gradient is performed with two consecutive propagations, one forward for the original system equations, and one backward in order to obtain $\hat{A}[u](t)$. These propagations provide the necessary ingredients to compute Eq. (15). In the next section, we will make a link of these forward and backward propagations to the formulation of the quantum dynamics via the Keldysh contour formalism.

It is also easy to see that all variations and generalizations of the QOCT equations naturally follow from our linear response approach.

A. Pure states

For the case of a pure-state dynamics, the density matrix takes the form $\hat{\rho}[u](t) = |\Psi[u](t)\rangle\langle\Psi[u](t)|$, where the wave function $|\Psi[u](t)\rangle$ evolves from a given initial state $|\Psi[u](t_0)\rangle = |\Psi_0\rangle$ according to the Schrödinger equation

$$\frac{\partial}{\partial t}|\Psi[u](t)\rangle = -i\hat{H}[u](t)|\Psi[u](t)\rangle. \tag{16}$$

The gradient of the merit function is given by the general Eq. (8). The only difference is that now the initial density matrix entering the response function describes a pure state $\hat{\rho}_0 = |\Psi_0\rangle\langle\Psi_0|$. Hence, Eq. (9) reduces to the form

$$\chi_{\hat{A},\hat{V}}(t,\tau) = -i\theta(t-\tau)\langle\Psi_0|[\hat{A}_H(t), \hat{V}_H(\tau)]|\Psi_0\rangle. \tag{17}$$

By inserting this equation into Eq. (8), writing the commutator explicitly, and inspecting the terms, we find that the gradient can be written as

$$\frac{\partial G}{\partial u}[u] = 2\,\mathrm{Im}\int_{t_0}^{t_f} d\tau\ \frac{\partial \epsilon}{\partial u}[u](\tau)\langle\chi[u](\tau)|\hat{V}|\Psi[u](\tau)\rangle, \tag{18}$$

where $|\chi[u](t)\rangle$ is defined by the expression

$$|\chi[u](t)\rangle = \hat{U}(t,t_f)\hat{A}|\Psi[u](t_f)\rangle. \tag{19}$$

Alternatively, this function can be viewed as a solution to the following backward propagation problem:

$$\frac{\partial}{\partial t}|\chi(t)\rangle = -i\hat{H}[u](t)|\chi(t)\rangle\,, \tag{20}$$

$$|\chi(t_f)\rangle = \hat{A}|\Psi[u](t_f)\rangle\,, \tag{21}$$

which coincides with the standard QOCT equations for pure states. Within the usual formalism, the state $|\chi[u](t)\rangle$ appears as a "Lagrange multiplier" wave function.

B. Continuous parameters

The case in which the control function $\epsilon(t)$ is not parametrized, but one does the search in the whole space of continuous functions, can also be treated essentially in the same manner. In this case, instead of a gradient, we will obtain a functional derivative; in fact, this derivative is nothing else than the response function, i.e., Eq. (8) is simply

$$\frac{\delta G}{\delta \epsilon(t)} = \chi_{\hat{A},\hat{V}}(t_f,t)\,. \tag{22}$$

This can be rewritten, for the pure state case, as

$$\frac{\delta G}{\partial \epsilon(t)} = 2\,\mathrm{Im}\langle\chi[\epsilon](t)|\hat{V}|\Psi[\epsilon](t)\rangle\,, \tag{23}$$

where $\chi[\epsilon](t)$ is the solution to Eqs. (20) and (21).

C. General target functionals

In some cases, the function to optimize is not a simple expectation value of an operator $\hat{A}$, but perhaps a more general expression in the form

$$G[u] = F[\hat{\rho}[u], u]\,, \tag{24}$$

where F is a functional of the evolution of the system (and also perhaps explicitly of the control parameters, hence, the second argument). Normally, this is split as

$$G[u] = J_1[\hat{\rho}[u]] + J_2[u]\,, \tag{25}$$

i.e., the first term is the *real* objective, depending on the evolution of the system, whereas the second term is added in order to penalize undesired features of the control function such as, for example, too high frequencies or intensities. In any case, any physically meaningful definition for J_1 will be that in which it is a function of expectation values of observables. In this case, the derivation outlined here is directly applicable by a simple use of the chain rule.

D. Time-dependent targets

A more interesting generalization is that in which the function to optimize depends on the expectation value of the operator at all times during the propagation, and not only at the final time t_f: Once again, this case can also be put in response-function language in a rather straightforward manner. Let us consider, for example, the pure-state case

$$G[u] = \int_{t_0}^{t_f} dt\ g(t)\langle\Psi[u](t)|\hat{A}|\Psi[u](t)\rangle\,, \tag{26}$$

where $g(t)$ is some weight function. The application of the LRT equations leads now to

$$\frac{\partial G}{\partial u}[u] = \int_{t_0}^{t_f} dt\ \int_{t_0}^{\infty} d\tau\ g(t)\frac{\partial \epsilon}{\partial u}[u](\tau)\chi_{\hat{A},\hat{V}}(t,\tau)\,. \tag{27}$$

Here, the response function $\chi_{\hat{A},\hat{V}}(t,\tau)$ is given by Eq. (17). Following the same route as in derivation of Eq. (18) in Sec. III A, we rewrite Eq. (27) as

$$\frac{\partial G}{\partial u}[u] = 2\,\mathrm{Im}\int_{0}^{t_f} d\tau\ \frac{\partial \epsilon}{\partial u}[u](\tau)\langle\chi[u](\tau)|\hat{V}|\Psi[u](\tau)\rangle, \tag{28}$$

FIG. 1. Keldysh contour.

where $\chi[u](\tau)$ is defined by the following integral:

$$|\chi[u](\tau)\rangle = \int_{\tau}^{t_f} dt\, g(t)\hat{U}(t,\tau)\hat{A}|\Psi[u](t)\rangle\,, \tag{29}$$

which can be put in the equivalent differential form

$$\frac{\partial}{\partial\tau}|\chi[u](\tau)\rangle = -i\hat{H}[u](\tau)|\chi[u](\tau)\rangle - g(\tau)\hat{A}|\Psi[u](\tau)\rangle, \tag{30}$$

$$|\chi[u](t_f)\rangle = 0\,. \tag{31}$$

These are once again the backward QOCT equations in the case of "time-dependent targets."

III. QOCT IN TERMS OF THE KELDYSH CONTOUR FORMALISM

The new point of view on QOCT proposed in the previous section naturally suggests new approximation strategies for control problems in interacting many-electron systems. As we will now show, the QOCT equations can be expressed in terms of correlations functions defined on a Keldysh [17] closed-time contour. This allows for an immediate application of the powerful machinery of nonequilibrium Green's function theory to the coherent control problem.

Let us reconsider the key equation for the gradient of the merit function [Eq. (8)] and write it explicitly as

$$\frac{\partial G}{\partial u}[u] = -i\int_{t_0}^{t_f} d\tau\, \frac{\partial\epsilon}{\partial u}[u](\tau)\mathrm{Tr}\{\hat{\rho}(t_0)\hat{A}_H(t_f)\hat{V}_H(\tau)\} - i\int_{t_f}^{t_0} d\tau\, \frac{\partial\epsilon}{\partial u}[u](\tau)\mathrm{Tr}\{\hat{\rho}(t_0)\hat{V}_H(\tau)\hat{A}_H(t_f)\}. \tag{32}$$

The two integrals in this equation can be composed into a single integral over the Keldysh contour C depicted in Fig. 1. This contour starts at t_0, goes froward in time to t_f, and then comes back to the origin. Therefore, by using the standard definition of a contour-ordered correlation function

$$\chi^C_{\hat{A},\hat{V}}(\tau,\tau') = -i\mathrm{Tr}\{\hat{\rho}(t_0)\mathrm{T}_C[\hat{A}_H(\tau)\hat{V}_H(\tau')]\}, \tag{33}$$

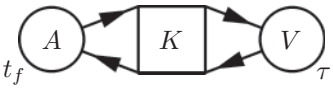
where T_C is the chronological ordering operator on the contour C, we can cast Eq. (32) into the following compact form:

$$\frac{\partial G}{\partial u}[u] = \int_C d\tau\, \frac{\partial\epsilon}{\partial u}[u](\tau)\chi^C_{\hat{A},\hat{V}}(t_f,\tau). \tag{34}$$

The main advantage of the representation (34) is that, for interacting many-body systems, the contour-ordered correlation functions can be calculated using the standard diagrammatic technique for nonequilibrium Keldysh Green's functions (see, e.g., Refs. [17,19,20,23–26]). In other words, by employing the well-developed machinery and approximations of the nonequilibrium Green's function theory (NEGFT), we can express the gradients of the merit function as a functional of the contour ordered one-particle Green's functions.

To illustrate the above statements, we consider the simplest situation when both the control field $\hat{V}$ and the observable of interest $\hat{A}$ are represented by one-particle operators. In this case, the correlation function $\chi^C_{\hat{A},\hat{V}}(t_f,\tau)$ entering Eq. (34) is given by

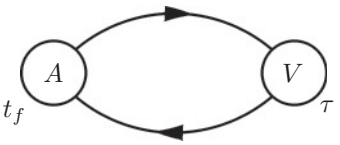
$$\chi^C_{\hat{A},\hat{V}}(t_f,\tau) =$$



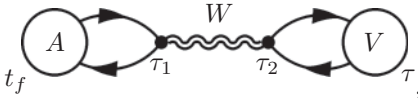
, (35)

where K is the exact two-particle Green's function. Now, we can take our favorite many-body approximation, such as Hartree-Fock, second-Born, T-matrix, random phase approximation (RPA), etc., to get an explicit and practically feasible expression. For example, at the RPA-GW level, the correlation function reduces to the two following terms:

$$\chi^C_{\hat{A},\hat{V}}(t_f,\tau) =$$



$$+$$



, (36)

Analytically, this diagram translates to

$$\begin{aligned}\chi^C_{\hat{A},\hat{V}}&(t_f,\tau)\\ &= \mathrm{Tr}\{\hat{A}G(t_f,\tau)\hat{V}G(\tau,t_f)\}\\ &\quad + \int d\tau_1 d\tau_2 \int d\mathbf{r_1} d\mathbf{r_2}\mathrm{Tr}\{\hat{A}G(t_f,\tau)\hat{n}(\mathbf{r_1})G(\tau,t_f)\}\\ &\quad \times W(\mathbf{r_1},\tau_1;\mathbf{r_2},\tau_2)\mathrm{Tr}\{\hat{n}(\mathbf{r_2})G(t_f,\tau)\hat{A}G(\tau,t_f)\},\end{aligned} \tag{37}$$

where $G(\tau_1,\tau_2) = G(\mathbf{r_1},\tau_1;\mathbf{r_2},\tau_2)$ is the one-particle contour Green's function, $W(\mathbf{r_1},\tau_1;\mathbf{r_2},\tau_2)$ is a dynamically screened Coulomb interaction, $\hat{n}(\mathbf{r})$ is a one-particle density operator, and all traces are taken over a one-particle Hilbert space.

Equation (37) shows that, for the practical calculation of the correlation function $\chi^C_{\hat{A},\hat{V}}(t_f,\tau)$, and thus the gradient of Eq. (34), we need the contour-ordered Green's function G and the screened interaction W. The latter is given by the RPA integral equation

, (38)

while the former is calculated by propagating the Kadanoff-Baym equation [19]

$$\left(i\frac{\partial}{\partial\tau_1} - \hat{h}(1)\right)G(1,2) = \delta(1,2) + \int d3\Sigma(1,3)G(3,2), \tag{39}$$

and its conjugate on the time contour. In Eq. (39), $\hat{h}(1) = \hat{h}(\mathbf{r_1},\tau_1)$ is the one-particle Hamiltonian, which also includes the Hartree potential, and the self-energy is given by the GW diagram

$$\Sigma(1,2) = G(1,2)W(2,1) =$$

, (40)

More technical details can be found, for example, in Ref. [25]. At this point, it is worth commenting on one technical issue. Most currently existing implementations of the Kadanoff-Baym equations [24–26] assume that the dynamics starts from the thermal equilibrium state at some temperature $T = 1/\beta$. The equilibrium initial conditions are technically convenient because they can be treated by a slight modification of the Keldysh contour. Namely, one attaches a "vertical track" going from t_0 to $t_0 - i\beta$ from the backward branch of the contour, and imposes antiperiodic Martin-Schwinger boundary conditions $G(t_0 - i\beta,\tau) = -G(t_0,\tau)$ on the Green's function. If this formalism is employed, then all time integrations in Eqs. (33) and (39) are along the modified contour including the vertical track. However, this does not influence the calculation of the gradient of Eq. (34) as it requires only the correlation function on the real-time forward and backward branches of the contour. We would like to emphasize that the use of equilibrium and ground-state initial conditions is not a fundamental restriction of NEGFT. It is also possible to formulate the theory for a general initial state [20,23,27,28], although we are not aware of any practical implementation of this formalism.

We conclude this section by noting the following remarkable fact regarding the Keldysh contour formulation of QOCT for interacting many-body systems. If the quantum dynamics is described within NEGFT, the implementation of QOCT does not require solving any additional equation. All ingredients required to calculate the merit-function gradients are already known from the solution of the Kadanoff-Baym equations. For example, at the RPA-GW level of the theory, one only needs to plug the known functions G and W into Eqs. (33) and (34), perform the integrations, and close the optimization loop.

IV. CONCLUSIONS

We have shown how the key equations of QOCT can be easily derived by employing the formalism of linear response theory. These equations provide the gradient of the target functional with respect to the external field, which has to be optimally shaped. In light of the linear response interpretation, the gradient is in fact the response function of the driven system. First of all, this derivation is valuable methodologically as it explains the internal structure of the coherent control theory using one of the most common techniques in theoretical physics, thus making QOCT more clear and accessible to a broad audience. In addition to that, our LRT representation immediately suggests a reformulation of QOCT equations in terms of the Keldysh contour-ordered correlation functions. The theory of nonequilibrium Green's functions (NEGFT) may then be directly applied to derive approximation strategies for control problem in interacting many-electron systems. Hence, our formalism establishes a connection of QOCT to the standard many-body perturbation theory and, thus, opens a way for solving various realistic coherent control problems at the *ab initio* level.

The implementation of QOCT looks especially simple and natural, if the quantum dynamics is described within NEGFT. In this case, the calculation of the merit-function gradients reduces to a straightforward integration, as all required quantities are already known from the solution of the Kadanoff-Baym equations. From the numerical point of view, solving the Kadanoff-Baym equations constitutes, in fact, the most time-consuming part of the optimization problem. Recently, several groups implemented the real-time propagation of the Kadanoff-Baym equations for many-electron dynamics in atoms and molecules [25,29], in strongly interacting nanoclusters [26,30], and for transient dynamics in the quantum transport situation [31]. The success of those implementations proves the feasibility of our approach to the many-body QOCT: the contour propagation schemes need only to be coupled to an optimization algorithm such as, for example, conjugate gradients. The full problem will require a number of gradient computations that will depend on the nature of the optimization problem and on the number of free parameters. Work on the numerical implementation of the Keldysh many-body QOCT proposed in this work is currently in progress. The results will be presented separately elsewhere.

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APPENDIX: GENERALIZED KUBO FORMULA

Let us consider a system governed by a total Hamiltonian $\hat{H}(t)$ that is split as

$$\hat{H}(t) = \hat{H}_0(t) + f(t)\hat{V}\,, \tag{A1}$$

given some real time-dependent function $f(t)$ supported in the time interval $[t_0,t_f]$. To formulate a generalized LRT, we need to solve the equation of motion for the density matrix $\hat{\rho}(t)$:

$$i\frac{\partial}{\partial t}\hat{\rho}(t) = [\hat{H}_0(t) + f(t)\hat{V},\hat{\rho}(t)] \tag{A2}$$

for some given initial $\hat{\rho}(t_0)$, by considering the second term as a "perturbation," while allowing the first term $\hat{H}_0$ to be time dependent.

We search for a solution in the form

$$\hat{\rho}(t) = \hat{\rho}_0(t) + \hat{\rho}_1(t), \tag{A3}$$

where $\hat{\rho}_0(t)$ solves Eq. (A2) with $f(t) = 0$ and the initial condition $\hat{\rho}_0(t_0) = \hat{\rho}(t_0)$, and $\hat{\rho}_1(t)$ is a solution to the linearized equation

$$i\frac{\partial}{\partial t}\hat{\rho}_1(t) = [\hat{H}_0(t),\hat{\rho}_1(t)] + [f(t)\hat{V},\hat{\rho}_0(t)] \tag{A4}$$

with the initial condition $\hat{\rho}_1(t_0) = 0$.

It is convenient to introduce a propagator $\hat{U}(t,t')$ for the unperturbed evolution

$$\hat{U}(t,t') = \hat{T}e^{-i\int_{t'}^{t} d\tau \hat{H}_0(\tau)}\,, \tag{A5}$$

where $\hat{T}$ is the usual time-ordering operator. Equation (A5) is a formal solution to the equations

$$\begin{aligned} i\frac{\partial}{\partial t}\hat{U}(t,t') &= \hat{H}_0(t)\hat{U}(t,t'), \\ i\frac{\partial}{\partial t'}\hat{U}(t,t') &= -\hat{U}(t,t')\hat{H}_0(t') \end{aligned} \tag{A6}$$

with the boundary condition $\hat{U}(t,t) = \hat{I}$.

Using Eqs. (A6), we immediately find both the unperturbed density matrix $\hat{\rho}_0(t)$ and the solution $\hat{\rho}_1(t)$ of the linearized equation (A4):

$$\hat{\rho}_0(t) = \hat{U}(t,t_0)\hat{\rho}(t_0)\hat{U}(t_0,t), \tag{A7}$$

$$\hat{\rho}_1(t) = -i\int_{t_0}^{t} d\tau\, \hat{U}(t,\tau)[f(\tau)\hat{V},\hat{\rho}_0(\tau)]\hat{U}(\tau,t). \tag{A8}$$

It is easy to check that $\hat{\rho}_1(t)$ of Eq. (A8) is the solution to Eq. (A4). Indeed, the differentiation with respect to the upper limit of the τ integral in Eq. (A8) yields the second term in the right-hand side in Eq. (A4), while the t derivatives of the propagators in Eq. (A8) produce the first term $[\hat{H}_0(t),\hat{\rho}_1(t)]$.

Now, one can calculate the change $\delta A(t)$ of the expectation value for any observable $\hat{A}$, which is induced by the perturbation [the second term in the Hamiltonian (A1)]

$$\delta A(t) = \mathrm{Tr}\{\hat{\rho}_1(t)\hat{A}\}. \tag{A9}$$

By inserting $\hat{\rho}_1(t)$ of Eq. (A8) into Eq. (A9) and rearranging terms, we get the result

$$\delta A(t) = -i\int_{t_0}^{t} d\tau\, f(\tau)\mathrm{Tr}\{\hat{\rho}(t_0)[\hat{A}_H(t),\hat{V}_H(\tau)]\}, \tag{A10}$$

where operators $\hat{O}(t)$ in the Heisenberg representation are defined as

$$\hat{O}_H(t) := \hat{U}(t_0,t)\hat{O}\hat{U}(t,t_0) \equiv \hat{U}^{\dagger}(t,t_0)\hat{O}\hat{U}(t,t_0). \tag{A11}$$

Equation (A10) suggests the definition of the $(\hat{A},\hat{V})$ response function as

$$\chi_{\hat{A},\hat{V}}(t,t') = -i\theta(t-t')\mathrm{Tr}\{\hat{\rho}(t_0)[\hat{A}_H(t),\hat{V}_H(t')]\} \tag{A12}$$

so that

$$\delta A(t) = \int_{t_0}^{\infty} d\tau\, f(\tau)\chi_{\hat{A},\hat{V}}(t,\tau)\,. \tag{A13}$$

The response function of Eq. (A13) has the standard form of Kubo's formula [32]. The only minor difference is that for a time-dependent unperturbed Hamiltonian $\hat{H}_0(t)$, the Heisenberg operators [Eq. (A11)] are defined via the time-ordered exponential of Eq. (A5).

Finally, we note that the QOCT equations can also be derived in yet another different but equivalent manner by making use of the following identity for the quantum mechanical propagator associated to a Hamiltonian that depends on a parameter λ:

$$\frac{\partial}{\partial\lambda}\hat{U}_{\lambda}(t_f,t_0) = -i\int_{t_0}^{t_f} dt\, \hat{U}^{\dagger}_{\lambda}(t,t_f)\frac{\partial\hat{H}_{\lambda}}{\partial\lambda}(t)\hat{U}_{\lambda}(t,t_0)\,. \tag{A14}$$

With this identity, it is straightforward to compute the derivative of

$$\begin{aligned} G[u] &= \langle\Psi[u](t_f)|\hat{A}|\Psi[u](t_f)\rangle \\ &= \langle\Psi_0|\hat{U}_u(t_0,t_f)|\hat{A}|\hat{U}_u(t_f,t_0)|\Psi_0\rangle\,, \end{aligned} \tag{A15}$$

where $\hat{U}_u(t,t_0)$ is the propagator determined by the Hamiltonian $\hat{H}[u](t)$.

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