

Time-dependent density-matrix functional in Liouville space and the optical response of many-electron systems

V. Chernyak and S. Mukamel

Department of Chemistry, University of Rochester, Rochester, New York 14727

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A time-dependent functional approach is developed for calculating the dynamical optical response and ground-state properties of nonrelativistic fermion systems. The functional (effective action) uses the particle-hole components of the reduced single-electron density matrix as variables and is constructed using the natural orbitals as a reference. It is defined on pairs of trajectories that constitute a Liouville space path in the phase space of the classical analog of the quantum system. A perturbative "semiclassical" calculation of the effective Liouville-space action is used to incorporate correlation effects in the linear optical response.

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I. INTRODUCTION

The reaction to external perturbations is an important tool in fundamental studies as well as practical applications of nonrelativistic many-electron systems [1]. The response provides a general probe of structural and dynamical relaxation properties of materials. In this paper we develop a theory for the optical response of many-electron systems that is based on a dynamical functional approach. (Although we focus our attention on optical response, the proposed theoretical approach can be directly applied to a more general response to an arbitrary external field.) To set the stage we first discuss the calculation of the optical response using existing functional theories. The nature of the response is specified by the choice of a set of operators coupled to the external field and by the set of observables. In the case of optical response the external electromagnetic field is coupled to the microscopic polarization [2]. In coherent optical response the observables are related to the expectation value of the polarization: the positions and widths of resonances observed in frequency domain coherent measurements provide information on the energies and lifetimes of excited states. The time domain coherent response reflects the dynamical properties of electronic and nuclear motions [3]. Relaxation that usually involves incoherent processes [4] can also be probed using the incoherent optical response, where the observables (e.g., fluorescence line shapes) are represented by expectation values of bilinear combinations of the polarization operator [5]. In the theory of coherent optical response, the expectation value of the polarization is usually expanded in powers of the external (or Maxwell) electromagnetic field. The expansion coefficients, known as the optical susceptibilities [6], are then calculated by expanding them in the global (many-body) electron eigenstates, which can be calculated using certain approximation schemes [7], and optical properties are related to the eigenvalues and to matrix elements of the dipole operator [6]. The off-resonant or static response (i.e., the response to a time-independent external field) is directly related to ground-state properties.

Many-body methods are based on the reduced description of quantum systems. They can be formulated in terms of equations of motion for selected observables, which are

closed by invoking specific approximations [3]. These equations, which deal with the observables directly, further suggest an oscillator (quasiparticle) picture of the optical response: the linear part of the equations describes the oscillators (quasiparticle normal modes) whereas the nonlinear terms represent anharmonicities (or quasiparticle scattering) that are responsible for the nonlinear optical response.

Equations of motion that are closed by using relatively simple factorization schemes provide a clear and simple picture of optical response in various materials. Equations for single-exciton, two-exciton, and exciton population variables in molecular systems [8,9], the Bloch-Maxwell equations in highly excited semiconductors [10], the time-dependent Hartree equations in metals and metal clusters [11], and the time-dependent Hartree-Fock (TDHF) equations [12] have been widely used to study the optical response mechanism. The TDHF equations, which can be applied to an arbitrary nonrelativistic (i.e., conserving the number of particles) system of interacting fermions, will play a central role in our theory. Many-body Green function techniques have some clear advantages, particularly for large systems where the response mostly reflects statistical properties such as level correlations that show up in multidimensional spectral densities, rather than properties of specific levels. These level correlations induce collective resonances in the nonlinear optical response [5]. The approach is much less demanding computationally and is size consistent, i.e., it properly describes the scaling of the nonlinear response with system size [8,13]. This has been demonstrated for molecular aggregates with confined Frenkel excitons [8]. There are two important issues in constructing a reduced description: the choice of variables and the way the equations are closed. There is no general way to address the first problem: the choice of variables depends on the properties of the system and requires good physical intuition since a proper choice can simplify the solution of the second problem. Once the variables are fixed, the second problem can be solved by deriving equations of motion for these variables. It is desirable to start with a formally exact universal equation for the externally driven quantum system. Practical applications can then be made by employing perturbative techniques. An important step towards the construction of a universal equation has been taken by Hohenberg and Kohn [14], who have introduced the den-

sity functional to study the ground state of a system of electrons. The procedure starts by introducing the ground-state energy $E[u(\mathbf{r})]$ as a functional of an external potential $u(\mathbf{r})$ [15]. $E[u(\mathbf{r})]$ is a generating functional for the static (off-resonant) response functions with respect to $u(\mathbf{r})$: the n -th-order response function is given by a $(n+1)$ -order variational derivative of the functional. Let $n(\mathbf{r})$ be the expectation value of the charge density and

$$F_0[u(\mathbf{r})] \equiv E[u(\mathbf{r})] + \int n(\mathbf{r})u(\mathbf{r})d\mathbf{r}. \quad (1.1)$$

The crucial step in the Hohenberg-Kohn procedure is to express $u(\mathbf{r})$ through $n(\mathbf{r})$ in Eq. (1.1), which leads to the density functional $F[n(\mathbf{r})]$ satisfying the equation $\delta F[n]/\delta n(\mathbf{r}) = u(\mathbf{r})$, which is the universal equation of the Hohenberg-Kohn theory (HKUE): it allows us to find the ground-state charge density of a system subjected to an external potential, provided the density functional is known. We can then solve the ground-state energy and calculate the static response. This procedure of calculating the response seems impractical at first; in the derivation of the HKUE we have expressed $u(\mathbf{r})$ through $n(\mathbf{r})$ by inverting the expression of $n(\mathbf{r})$ in terms of $u(\mathbf{r})$. In other words, we apparently need to know the response functions to start with. However, one never uses this scheme to calculate $F[n]$; the derivation is merely needed to prove that such a functional and hence the HKUE do exist. In practice, $F[n]$ is calculated using other techniques. For example, one can use a perturbation theory in a small expansion parameter to express $n(\mathbf{r})$ through $u(\mathbf{r})$ in first order. By comparing this expression with the HKUE it is possible to calculate $F[n]$ to first non-vanishing order. If one now solves the HKUE using $F[n]$ obtained this way, the solution $n(\mathbf{r})$ will be different from the expression of $n(\mathbf{r})$ through $u(\mathbf{r})$ used to obtain $F[n]$, since the latter is only the first-order expansion of the solution $n(\mathbf{r})$ in the small parameter rather than the solution itself. Therefore, this procedure leads to the expansion of the equation and gives a perturbative way to evaluate the density functional. *Ab initio* calculations of the density functional usually use an expansion in two parameters: inverse density n^{-1} and density gradients ∇n [16].

A different procedure introduced by Kohn and Sham [17] is used in most applications of the Hohenberg-Kohn theory. In the Kohn-Sham approach the exact ground-state density $n(\mathbf{r})$ is represented as the density of an effective system of noninteracting particles in an effective potential $u_{\text{eff}}(\mathbf{r}, [n])$, which is a functional of the density. The Hohenberg-Kohn theory is then used to obtain an exact relation between $F[n]$ and $u_{\text{eff}}(\mathbf{r}, [n])$. The basic equation of the Kohn-Sham theory is $n(\mathbf{r}) = n_0(\mathbf{r}, [u_{\text{eff}}[n]])$, where $n_0(\mathbf{r}, [u])$ is the density of noninteracting particles in the potential $u(\mathbf{r})$. Kohn and Sham further represented the right-hand side (rhs) of their equation in terms of effective orbitals, i.e., the single-electron states in the potential $u_{\text{eff}}(\mathbf{r})$. A scheme that does not require effective orbitals has been proposed by Yang [18], who has used the path-integral representation of the rhs of the Kohn-Sham equation. His approach can be considered a variant of the Kohn-Sham theory since it uses a different form of the same equation.

The original equations of density-functional theory were static and the theory only deals with the equilibrium ground-state properties. The first important contribution to the time-dependent density-functional theory that allows one to treat the dynamical response was made by Peuckert [19]. Making use of the variational principle for the time-dependent Schrödinger equation, he showed that if the potential $u(\mathbf{r}, \tau)$ depends on time, the time-dependent density $n(\mathbf{r}, \tau)$ can be described by a system of noninteracting particles driven by the time-dependent effective potential $u_{\text{eff}}(\mathbf{r}, \tau, [n])$, which is a functional of $n(\mathbf{r}, \tau)$. A formal expression for the effective potential has been derived. Generalizations of the Hohenberg-Kohn theorem to the time-dependent case have been considered by several authors [20–22]. The time-dependent density-functional approach has been used successfully to study the dynamical linear [23–25] as well as the nonlinear [26] response. The dynamical equations for $n(\mathbf{r}, \tau)$ are usually formulated by introducing the effective time-dependent orbitals of a reference system of noninteracting particles driven by the effective potential. An approach that uses the path-integral instead of the orbital representation for the density of the effective system has been proposed by Yang [27].

Time-dependent density-functional theory (including the approach of [27]) generalizes the Kohn-Sham (rather than the Hohenberg-Kohn) approach since it deals with the effective potential. Quantum field theory [28] offers a particularly appealing generalization of the Hohenberg-Kohn density-functional approach to the time-dependent case, which allows an arbitrary choice of variables. The basic quantities in that approach are the time-ordered Green functions defined as expectation values with respect to the ground state of products of chronologically ordered Heisenberg operators. The resonances of the Green function in the frequency domain are related to particles. The following procedure is used to calculate these Green functions. Let $u(\mathbf{r}, \tau)$ be a time-dependent potential, $\hat{H}(t; [u])$ the time-dependent Hamiltonian, and $\hat{U}(t, t'; [u])$ the evolution operator:

$$\frac{\partial \hat{U}(t, t'; [u])}{\partial t} = -i\hat{H}(t; [u])\hat{U}(t, t'; [u]), \quad (1.2a)$$

$$\hat{U}(t', t'; [u]) = \hat{I}. \quad (1.2b)$$

We next introduce the generating function $S_0[u]$, defined by

$$\exp\{iS_0[u(\mathbf{r}, t)]\} \equiv \langle \Omega_0 | \hat{U}(\infty, -\infty; [u(\mathbf{r}, \tau)]) | \Omega_0 \rangle, \quad (1.3)$$

where $|\Omega_0\rangle$ is the ground state. The time-dependent analog of the Hohenberg-Kohn functional is denoted the effective action S_{eff} and has the form

$$S_{\text{eff}}[n(\mathbf{r}, \tau)] \equiv S_0[u(\mathbf{r}, \tau)] - \int d\mathbf{r} d\tau u(\mathbf{r}, \tau)n(\mathbf{r}, \tau), \quad (1.4)$$

where $u(\mathbf{r}, \tau)$ should be expressed in terms of $n(\mathbf{r}, \tau)$. The basic equation now becomes

$$\frac{\delta S_{\text{eff}}[n]}{\delta n(\mathbf{r}, \tau)} = -u(\mathbf{r}, \tau). \quad (1.5)$$

In analogy with the density-functional calculations, S_{eff} should be evaluated using a different, independent, route.

As can be seen from Eq. (1.3), since the evolution operator has a form of a time-ordered exponent ($\hat{U}(t, t') = T \exp[-i \int_{t'}^t \hat{H}(\tau) d\tau$], where T is the operator of chronological ordering), $S_0[u(\mathbf{r}), t]$ is the generating functional for time-ordered Green functions. This means that the effective action formalism can be used to obtain the time-ordered Green functions; however, the nonlinear response is expressed in terms of nonequilibrium (Liouville space or Keldysh-Schwinger) [29] Green functions [5,30] and this action is not sufficient for calculating the dynamical optical response.

A formal Liouville space [31] approach to quantum statistical mechanics has been recently developed [32-34]. The theory is based on introducing operators acting in Liouville space with a double number of variables (twin operators). A nonequilibrium time-dependent functional theory in the Liouville space can be obtained by combining the ideas of Liouville quantum dynamics and the quantum-mechanical dynamical variational principle [35,36]. A broader set of variables for the argument of the functional (including the expectation values of the current, electromagnetic field, and variables responsible for ionic motions) has been introduced as well.

Constructing the functional is the most challenging problem in the application of density-functional theory. The common procedure involves a gradient expansion, taking into account only the lower gradients [16] and making an ansatz on the dependence of the coefficients on local density. In the time-dependent scheme gradients should include time derivatives and gradient expansions always fail near resonance; a completely different procedure for computing the effective action is needed.

In this paper we develop a generalized functional theory for calculating the linear and the nonlinear optical response of many electron systems. The approach applies to diverse systems such as molecular aggregates, conjugated polymers, and semiconductor nanoparticles. An important aspect of the present approach is the choice of an alternative set of variables, i.e., the particle-hole components of the reduced single-electron density matrix, as opposed to the charge density, as the argument of our functional. This yields the Liouville space Green functions necessary for computing the optical response. The choice of the charge density as the argument of the functional leads to a simple form of the functional only in systems that can be described by collective charge-density variables (e.g., metals). Systems with strong electron correlations (such as conjugated polymers) can, in principle, be described in the framework of density-functional theory; however, the density functional in the relevant range of densities becomes very complicated. For such systems the reduced single-electron density matrix [37], which describes the dynamics of electron-hole pairs, seems to be a more relevant variable (as opposed to the charge density, which includes only diagonal elements of the density matrix). The present generalization of time-dependent density-functional theory is based on the reduced electron density matrix as the argument of the functional. We represent the reduced density matrix in the presence of the external field as $\rho = \rho^{(g)} + \delta\rho$. The ground-state reduced density

matrix $\rho^{(g)}$ is assumed to be given. One can use the ground state obtained from either standard quantum chemistry calculations or the density-functional approach. We can then define the hole and the particle subspace as the subspace generated by the eigenvectors of $\rho^{(g)}$, known as the natural orbitals [37-39] and its orthogonal components, respectively. We have chosen the particle-hole components of $\delta\rho$ as the variables of the effective action. The operators corresponding to the other components can be expressed in terms of these particle-hole variables and, consequently, can be calculated provided the effective action is known. The reason for our choice of variables is as follows. We shall show that a system of TDHF equations can be associated with any quantum nonrelativistic fermion system and these equations can be treated as classical Hamiltonian dynamical equations on the phase space consisting of the set of all single Slater determinants [40]. Proper geometrical quantization [41] of this classical dynamical system brings us back to the original quantum system. In the vicinity of equilibrium, the Slater determinants can be described by the particle-hole components of related reduced density matrices. It makes sense to choose the variables of the effective action to be related to the variables of the classical analogue of the system. This enables us to treat derivations from the TDHF approximation as "semiclassical" corrections. Note that this semiclassical expansion has nothing to do with \hbar . The expansion parameter is rather connected with the strength of correlations in the system.

An additional ingredient of the present approach is that we formulate the theory in a way that makes it possible to follow the evolution of general (nonlinear) functions of the functional arguments. This gives more flexibility in applications of time-dependent functional theory to the theory of the optical response. In particular, one can consider external fields that are coupled to nonlinear combinations of the functional arguments. The time-dependent functional theory based on the variational principle allows us to follow only the quantum evolution of linear combinations of the functional variables. This extension is essential in order to calculate the optical response with respect to a field coupled to all components of the reduced density matrix, while using only the particle-hole components as the function arguments. We will also adopt a path-integral definition of the effective action as is common in field theory [28]. We further develop a perturbative expansion for the functional and derive closed expressions for the linear response. The extension to the nonlinear response is straightforward.

In Sec. II we express the optical response of a quantum system in terms of nonequilibrium Green functions of the polarization operators. We then introduce the effective action in Liouville space with respect to a set of operators \hat{X} and reduce the problem of calculating the response to solving equations of motion that are determined by the effective action. In Sec. III we develop a perturbation theory for the effective action, which allows us to calculate the optical response perturbatively starting with a zeroth-order Hamiltonian that is not a free-particle Hamiltonian. In Sec. IV we apply the results of Secs. II and III to a general model of nonrelativistic interacting fermions, choosing the particle-hole components of the reduced single-electron density matrix as the arguments of the effective action. In Sec. V we

calculate perturbatively the quadratic terms of the effective action and obtain expressions for the linear response connected with the particle-hole components of polarization. In Sec. VI we obtain an eigenmode expansion of the linear response that is very convenient for describing its resonant properties. In Sec. VII we evaluate the contributions to the linear response connected with the particle-particle and hole-hole components of the density matrix. Finally, in Sec. VIII we show how the effective action formalism can be applied to calculate ground-state properties.

II. OPTICAL RESPONSE AND THE EFFECTIVE ACTION IN LIOUVILLE SPACE

In this section we connect the optical response to correlation functions in Liouville space (Keldysh-Schwinger Green functions) and express the latter in terms of the effective action. Consider a quantum system described by the Hamiltonian

$$\hat{H}_T(t) = \hat{H} - \mathcal{E}(t) \cdot \hat{P}. \quad (2.1)$$

Here \hat{H} is the Hamiltonian of the isolated system and \hat{P} denotes a set of operators interacting with a set of external fields $\mathcal{E}(t)$. We introduce the evolution operator $\hat{U}(t, t')$ for the isolated system

$$\frac{d\hat{U}(t, t')}{dt} = -i\hat{H}\hat{U}(t, t'), \quad (2.2)$$

with $\hat{U}(t', t') = \hat{I}$. In the interaction picture, the time evolution of an arbitrary operator \hat{Q} is given by $\hat{Q}(t) \equiv \hat{U}^\dagger(t, -\infty)\hat{Q}\hat{U}(t, -\infty)$. Following [5,30] we introduce chronological Green functions on the Keldysh time loop [29]. For operators $\hat{Q}^{(1)}, \dots, \hat{Q}^{(n)}$ we define the chronological correlation function $\langle \hat{Q}_{i_1}^{(1)}(t_1) \dots \hat{Q}_{i_n}^{(n)}(t_n) \rangle$ with indices i_1, \dots, i_n which assume the values $i = L, R$:

$$\begin{aligned} & \langle \hat{Q}_L^{(1)}(t_1) \dots \hat{Q}_L^{(j)}(t_j) \hat{Q}_R^{(j+1)}(t_{j+1}) \dots \hat{Q}_R^{(n)}(t_n) \rangle \\ & \equiv \text{Tr}\{T[\hat{Q}^{(1)}(t_1) \dots \hat{Q}^{(j)}(t_j)]\hat{\rho}_M \\ & \quad \times T'[\hat{Q}^{(j+1)}(t_{j+1}) \dots \hat{Q}^{(n)}(t_n)]\}. \end{aligned} \quad (2.3)$$

The indices L and R indicate whether the operator should appear to the left or to the right of the equilibrium many-body density matrix $\hat{\rho}_M$. T stands for chronological time ordering operator. It acts on the "left" operators $\hat{Q}^{(1)}(t_1), \dots, \hat{Q}^{(j)}(t_j)$ and rearranges them so that the earliest time appears on the right and the latest time on the left. The operator T' stands for antichronological time ordering. It acts on the "right" operators $\hat{Q}^{(j+1)}(t_{j+1}), \dots, \hat{Q}^{(n)}(t_n)$ and rearranges them such that the earliest time is to the left and the latest time to the right. Since the order of operators is uniquely determined by the L and R indices and by the operators T and T' , the chronological correlation function is invariant to the ordering of operators on the left-hand side (lhs) of Eq. (2.3). We next change variables and define $Q_+ \equiv \frac{1}{2}(Q_L + Q_R)$ and $Q_- \equiv Q_L - Q_R$. We will label these variables Q_α , where the greek index α assumes the values $+$ and $-$. Using this notation, the expectation value of the

polarization in the extremely driven system is given by $P(t) = \langle \hat{P}_+(t) \exp\{i \int d\tau \mathcal{E}(\tau) P_-(\tau)\} \rangle$ [5,30]. The response function $R^{(n)}$ is introduced by expanding the expectation value of the polarization in powers of the external field

$$\begin{aligned} P(\tau) = & \sum_{n=0}^{\infty} \frac{1}{n!} \int d\tau_1 \dots d\tau_n R^{(n)}(\tau, \tau_1, \dots, \tau_n) \\ & \times \mathcal{E}(\tau_1) \dots \mathcal{E}(\tau_n), \end{aligned} \quad (2.4a)$$

with

$$R^{(n)}(\tau, \tau_1, \dots, \tau_n) = i^n \langle \hat{P}_+(\tau) \hat{P}_-(\tau_1) \dots \hat{P}_-(\tau_n) \rangle. \quad (2.4b)$$

The goal of any theory of optical response is to either calculate the polarization $P(\tau)$ directly or evaluate the response functions $R^{(n)}$ to a given order in the driving field.

The main objective of the effective action formalism in Liouville space is to develop a reduced description of complex many-body systems by following the time evolution of some selected dynamical variables of interest. An effective action is constructed that contains all the information necessary for calculating the expectation values and nonequilibrium (Keldysh) correlation functions of these dynamical variables. Using this approach we can formulate the optical response of a driven quantum system in a way that closely resembles the classical theory of optical response. Let us consider a classical system with a set of coordinates X . Within the Lagrangian formalism the system is described by its action $S[X(\tau)]$. The total action of the system driven by an external time-dependent field $h(\tau)$ is

$$S_T[X(\tau)] \equiv S[X(\tau)] - \int d\tau h(\tau) X(\tau). \quad (2.5)$$

Equations of motion are obtained by minimizing this action $\delta S_T / \delta X(\tau) = 0$ or, taking into account Eq. (2.5), $\delta S / X(\tau) = h(\tau)$.

Consider now a quantum system with a Hamiltonian \hat{H} . We introduce a pair of time-dependent external fields $h_j(\tau)$ and define the Hamiltonians $\hat{H}_j(\tau)$

$$\hat{H}_j(\tau) \equiv \hat{H} + h_j(\tau) \hat{X}, \quad j = L, R. \quad (2.6a)$$

The corresponding evolution operators $\hat{U}_j(t, t')$ satisfy the equations

$$\frac{d\hat{U}_j(t, t')}{dt'} = -i\hat{H}_j(t) \hat{U}_j(t, t'), \quad (2.6b)$$

with the initial conditions $\hat{U}_j(t', t') = \hat{I}$, where \hat{I} is the unit operator. \hat{X} is a set of operators and $\hat{X}(\tau)$ are the Heisenberg operators related to \hat{X} . It should be noted that the second term in Eq. (2.6a) does not necessarily represent a coupling with an actual physical field. \hat{X} is the set of operators for which we wish to develop a reduced description and $h(\tau)$ is in general a set of corresponding (fictitious) fields introduced in order to construct the reduced description. The connection to the actual external field and the desired response functions [Eq. (2.4b)] will be established at the end of this section.

We next define an action $S_0 = [\tilde{h}(\tau)]$ with $\tilde{h}(\tau) \equiv (h_L(\tau), h_R(\tau))$ by the equation

$$\exp\{iS_0[\tilde{h}(\tau)]\} \equiv \text{Tr}\{U_L(\infty, -\infty)\hat{\rho}_M U_R^\dagger(\infty, -\infty)\}, \quad (2.7)$$

where $\hat{\rho}_M$ is the equilibrium density matrix of the system. To express Eq. (2.7) in the path-integral form [42,43], we introduce a classical action $S[q(\tau)]$, which describes the classical analog of the original quantum system, where q stands for the complete set of classical variables. Equation (2.7) can be represented in terms of a path integral over trajectories in Liouville space $\tilde{q}(\tau)$, defined as pairs of trajectories [5,30] $\tilde{q}(\tau) = (q_L(\tau), q_R(\tau))$, with $q_L(\tau = +\infty) = q_R(\tau = +\infty)$. Introducing for an arbitrary function of q , $f(q)$, $f_-(\tilde{q}) \equiv f(q_L) - f(q_R)$, and $f_+(\tilde{q}) \equiv \frac{1}{2}[f(q_L) + f(q_R)]$, we obtain

$$\begin{aligned} \exp\{iS_0[\tilde{h}(\tau)]\} = & \int D[\tilde{q}(\tau)] \exp\left\{iS^{(k)}[\tilde{q}(\tau)] \right. \\ & \left. - i \int [h_+(\tau)X_-(\tau) + h_-(\tau)X_+(\tau)]d\tau\right\}, \end{aligned} \quad (2.8a)$$

with

$$S^{(k)}[\tilde{q}(\tau)] \equiv S[q_L(\tau)] - S[q_R(\tau)], \quad (2.8b)$$

and X is the set of classical variables related to the operators \hat{X} . The "expectation values" $x_\alpha(t)$, $\alpha = +, -$, are given by

$$\begin{aligned} x_\alpha(t) \equiv & \exp\{-iS_0[\tilde{h}(\tau)]\} \int D[\tilde{q}(\tau)] X_\alpha(t) \exp\{iS^{(k)}[\tilde{q}(\tau)]\} \\ & \times \exp\left\{-i \int d\tau [h_+(\tau)X_-(\tau) + h_-(\tau)X_+(\tau)]\right\}. \end{aligned} \quad (2.9)$$

We define the effective action $S_{\text{eff}}[x_\alpha(\tau)]$

$$\begin{aligned} S_{\text{eff}}[x_\alpha(\tau)] \equiv & S_0[\tilde{h}(\tau)] \\ & + \int d\tau [h_+(\tau)x_-(\tau) + h_-(\tau)x_+(\tau)], \end{aligned} \quad (2.10)$$

and $\tilde{h}(\tau)$ should be expressed in terms of $x_\alpha(\tau)$ in the rhs of Eq. (2.10) using Eq. (2.9). Using the effective action we can write the universal equations of motion for $x_+(\tau)$ and $x_-(\tau)$

$$\frac{\delta S_{\text{eff}}}{\delta x_-(\tau)} = h_+(\tau), \quad \frac{\delta S_{\text{eff}}}{\delta x_+(\tau)} = h_-(\tau). \quad (2.11)$$

Assuming that S_{eff} is known, we now show how it may be used for calculating the optical response. We shall distinguish between two cases. We first assume [case (i)] that the external field is coupled to the operators whose expectation values form the argument of S_{eff} . This means that the fictitious external field that is used to define S_{eff} coincides with the physical external field. In this case the connection to classical response theory is straightforward: We simply re-

place the operators \hat{X} by the polarization \hat{P} . The expectation value of \hat{X} in the presence of the driving field $h(\tau)$ can be obtained from Eq. (2.6b) with $h_L(\tau) = h_R(\tau) = h(\tau)$, which implies $h_-(\tau) = 0$, $h_+(\tau) = h(\tau)$. Denoting by $x(\tau)$ the expectation value of $\hat{X}(\tau)$ in the system driven by the external field $h(\tau)$, we obtain $x_-(\tau) = 0$, $x_+(\tau) = x(\tau)$. Substituting this into Eq. (2.11), we obtain

$$\left. \frac{\delta S_{\text{eff}}}{\delta x_-(\tau)} \right|_{x_-(\tau)=0, x_+(\tau)=x(\tau)} = h(\tau). \quad (2.12)$$

Solving Eq. (2.12) for $x(\tau)$ we can calculate the expectation value of $\hat{X}(\tau)$ in terms of the external field. This means that the reduced dynamics of a quantum system can be obtained nonperturbatively through a classical procedure of solving an equation of motion [Eq. (2.12)], and all quantum effects are incorporated in the renormalized classical action S_{eff} . In addition, we can obtain the response functions $R^{(n)}$ by solving Eq. (2.12) perturbatively in $h(\tau)$.

In many cases it is more convenient to define the variables in S_{eff} in such a way that the physical external field is coupled to some nonlinear functions of these variables. For example, in the application to fermion systems, discussed in Sec. IV, the external electric field is coupled to the reduced density matrix $\hat{\rho}_{\bar{m}\bar{n}} \equiv \hat{c}_m^\dagger \hat{c}_{\bar{n}}$. However, we shall choose only its particle-hole components as the effective action variables. The other components of $\hat{\rho}$ can be represented as commutators of the particle-hole components; on the operator level they are bilinear functions of the effective action variables. In this [case (ii)] the expectation values of \hat{X} [Eq. (2.12)] do not have a simple physical meaning since they correspond to a system driven by fictitious (nonphysical) fields. However, the correlation functions of \hat{X} can still be used to construct the optical response function. One needs first to express the optical susceptibilities in terms of correlation functions of the effective action variables in Liouville space and then calculate these correlation functions using the effective action. We will apply the following procedure. Let the physical external field be coupled to a set of polarization operators denoted by \hat{P} . The variables of S_{eff} are formed by expectation values of a set of operators denoted \hat{X} . We assume that \hat{P} are polynomial functions of \hat{X} , i.e., the external field is coupled to nonlinear functions of the effective action variables. The necessary correlation functions of \hat{P} can then be calculated by expressing the operators \hat{P} in terms of \hat{X} to obtain the response as a sum of Liouville-space correlation functions of \hat{X} . The Liouville-space correlation functions of \hat{X} in a system with no driving field can be expressed in terms of the effective action by applying the standard procedure (see [28]) to the path-integral representation on the Keldysh time loop [Eq. (2.9)], which is based on Eqs. (2.11). The result can be formulated as follows. Consider an auxiliary quantum system whose classical action is S_{eff} . Representing the effective action in a form

$$S_{\text{eff}} = S_{\text{eff}}^{(0)} + S_{\text{eff}}^{(\text{int})}, \quad (2.13)$$

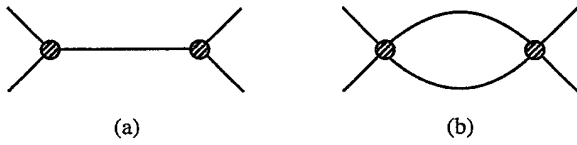


FIG. 1. Typical diagrams contributing to the Green functions: (a) not containing loops and contributing to Eqs. (2.14) and (2.15) and (b) containing a loop and contributing to Eq. (2.14) only.

where $S_{\text{eff}}^{(0)}$ contains the quadratic and $S_{\text{eff}}^{(\text{int})}$ the higher-order terms (the linear terms vanish), we can write the path-integral representation for the correlation functions in the new system in the form

$$\langle X_{\alpha_1}(\tau_1) \cdots X_{\alpha_n}(\tau_n) \rangle_{S_{\text{eff}}} = \langle X_{\alpha_1}(\tau_1) \cdots X_{\alpha_n}(\tau_n) \exp\{iS_{\text{eff}}^{(\text{int})}[X(\tau)]\} \rangle_{S_{\text{eff}}^{(0)}}. \quad (2.14)$$

The rhs of Eq. (2.14) can be evaluated by expanding the exponent and using Wick's theorem [42,44]. Each term in the expansion can be represented with a diagram (Fig. 1), where the vertices stand for the coefficients in the expansion of S_{eff} in powers of X and the lines denote the bare (with respect to $S_{\text{eff}}^{(0)}$) two-point Green functions. We denote by $\langle \rangle_{S_{\text{eff}}^{(0)}}^c$ the sum of all contributions represented by diagrams without loops. We then have for the Green functions in the original system (denoted $\langle \rangle_S$, since they are calculated with respect to the action S)

$$\langle X_{\alpha_1}(\tau_1) \cdots X_{\alpha_n}(\tau_n) \rangle_S = \langle X_{\alpha_1}(\tau_1) \cdots X_{\alpha_n}(\tau_n) \exp\{iS_{\text{eff}}^{(\text{int})}[X(\tau)]\} \rangle_{S_{\text{eff}}^{(0)}}^c, \quad (2.15)$$

which is the desired closed expression. Note that since we need to take into account only tree diagrams (i.e., diagrams without loops), each Green function is given by a finite number of contributions.

In summary, we have shown how the expectation value and correlation functions of the chosen set of variables \hat{X} may be obtained using the effective action. The expectation values are obtained by solving renormalized classical equations of motion [Eq. (2.12)]. The procedure of obtaining the correlation functions from S_{eff} may also be reduced to a classical procedure, i.e., solving classical equations of motion obtained by applying a variational procedure to S_{eff} . It therefore has the same level of complexity as solving the renormalized equations of motion for the expectation values.

III. PERTURBATIVE EXPANSION OF THE EFFECTIVE ACTION

The success of the effective action approach depends on a perturbative expansion of the action. Let \hat{H} be the Hamiltonian of a quantum system that can be partitioned as

$$\hat{H} = \hat{H}_0 + \hat{H}_1, \quad (3.1)$$

and $S_{\text{eff}}^{(0)}$ the effective action determined by the zeroth-order Hamiltonian \hat{H}_0 . Our goal is to obtain the effective action S_{eff} of the Hamiltonian \hat{H} in the form of an expansion in the small parameters of \hat{H}_1 . To that end, we partition the classical action of our quantum system $S = S_0 + S_1$, where S_0 and S_1 are related to \hat{H}_0 and \hat{H}_1 . Using the path-integral representation we obtain

$$\langle X_{\alpha_1}(\tau_1) \cdots X_{\alpha_n}(\tau_n) \rangle_S = \langle X_{\alpha_1}(\tau_1) \cdots X_{\alpha_n}(\tau_n) \exp\{iS^{(1)}[X(\tau)]\} \rangle_{S_0}. \quad (3.2)$$

Perturbation theories for Green functions are usually developed using Eq. (3.1) when \hat{H}_0 is a free-particle Hamiltonian, by expanding the exponent in Eq. (3.2) and using the Wick theorem [42,44]. A perturbation theory which applies when H_0 is not a free-particle Hamiltonian has been developed for spin systems by Vaks, Larkin, and Pikin [45]. Here we generalize that approach to arbitrary Hamiltonians in Liouville space and formulate the perturbation theory using the effective action language.

Expanding the exponent in the rhs of Eq. (3.2) we can, instead of using the Wick theorem (which does not apply in our case), use the Green-function expressions of Sec. II [Eq. (2.15)]. These express the correlation functions obtained on the rhs of Eq. (3.2) after the expansion in terms of the effective action $(S_0)_{\text{eff}}$ of \hat{H}_0 . Applying Eq. (2.15) for $S = S_0$, we obtain a diagrammatic perturbation theory. To introduce the vertices and the bare Green functions we present $(S_0)_{\text{eff}}$ and S_1 using the expansions

$$(S_0)_{\text{eff}}[x_\alpha(\tau)] = \sum_{n=2}^{\infty} \frac{1}{n!} \sum_{\alpha_1 \cdots \alpha_n} \int d\tau_1 \cdots d\tau_n \times S_{0\alpha_1 \cdots \alpha_n}^{(n)}(\tau_1, \dots, \tau_n) x_{-\alpha_1}(\tau_1) \cdots x_{-\alpha_n}(\tau_n), \quad (3.3a)$$

$$S_1[X_\alpha(\tau)] = \sum_{n=1}^{\infty} \frac{1}{n!} \sum_{\alpha_1 \cdots \alpha_n} \int d\tau_1 \cdots d\tau_n \times S_{1\alpha_1 \cdots \alpha_n}^{(n)}(\tau_1, \dots, \tau_n) X_{-\alpha_1}(\tau_1) \cdots X_{-\alpha_n}(\tau_n). \quad (3.3b)$$

In our diagram technique we have two types of vertices: (i) $S_{1\alpha_1 \cdots \alpha_n}^{(n)}(\tau_1, \dots, \tau_n)$, obtained by expanding the exponent in Eq. (3.2), and (ii) $S_{0\alpha_1 \cdots \alpha_n}^{(n)}(\tau_1, \dots, \tau_n)$, which appear in the process of evaluating the correlation functions after expanding the exponent, by applying the procedure of Sec. II. Since only tree diagrams contribute in Eq. (2.15), for calculating $\langle X_{\alpha_1}(\tau_1) \cdots X_{\alpha_n}(\tau_n) \rangle_S$ [see Eq. (3.2)] we take into account diagrams with both types of vertices, but apply a constraint: a diagram should not contain loops with only vertices of type (ii) [loops that contain at least one type (i)

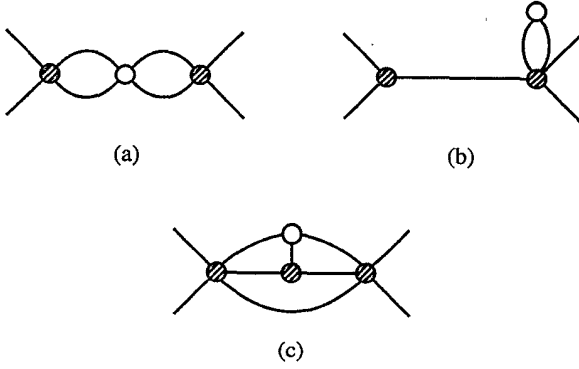


FIG. 2. Diagrams illustrating the diagram technique rules given at the end of Appendix B: (a) contributing to the Green function and to the effective action since loops contain a type-(i) vertex, (b) one-particle reducible contributing to a Green function but not to the effective action, and (c) contributing neither to the Green function nor to the effective action since it contains a loop involving type-(ii) vertices only. Open and hatched circles denote the type-(i) and type-(ii) vertices, respectively.

vertex are allowed]. The bare Green function $G^{(0)}$, which is represented by a line in the diagrams, can be found from the equation

$$\sum_{\beta=\pm} \int d\tau S_{0\alpha\beta}^{(2)}(\tau_1, \tau) G_{-\beta\alpha'}^{(0)}(\tau, \tau_2) = \delta_{\alpha, -\alpha'} \delta(\tau_1 - \tau_2). \quad (3.4)$$

The diagrammatic rules for the correlation functions are as follows.

- (1) A correlation function is given by a sum of contributions represented by diagrams.
- (2) Lines in the diagrams are related to $G^{(0)}$, which can be found from Eq. (3.4).
- (3) There are two types of vertices:
- (i) $S_{1\alpha_1 \dots \alpha_n}^{(n)}(\tau_1, \dots, \tau_n)$ and (ii) $S_{0\alpha_1 \dots \alpha_n}^{(n)}(\tau_1, \dots, \tau_n)$.
- (4) Diagrams should contain loops involving type-(ii) vertices only.

We can now formulate a perturbation theory for the effective action since we have a perturbation theory for the correlation functions, which in turn can be expressed as a sum of tree diagrams with vertices representing the expansion of S_{eff} :

$$S_{\text{eff}}[x_\alpha(\tau)] = \sum_{n=2}^{\infty} \frac{1}{n!} \sum_{\alpha_1 \dots \alpha_n} \int d\tau_1 \dots d\tau_n \times S_{\alpha_1 \dots \alpha_n}^{(n)}(\tau_1, \dots, \tau_n) x_{-\alpha_1}(\tau_1) \dots x_{-\alpha_n}(\tau_n). \quad (3.5)$$

The rules of the perturbation theory for S_{eff} are as follows.

- (1) $S_{\alpha_1 \dots \alpha_n}^{(n)}(\tau_1, \dots, \tau_n)$ is given as the sum of terms represented by diagrams with n external ends.
- (2) Lines in diagrams represent $G^{(0)}$ from Eq. (3.4).
- (3) There are two types of vertices:
- (i) $S_{1\alpha_1 \dots \alpha_n}^{(n)}(\tau_1, \dots, \tau_n)$ and (ii) $S_{0\alpha_1 \dots \alpha_n}^{(n)}(\tau_1, \dots, \tau_n)$.

(4) The diagrams should be connected and one-particle irreducible. The latter condition implies that (a) a diagram does not contain the factors $G^{(0)}$ related to external lines and (b) a diagram cannot be cut by one line into two disconnected parts.

(5) The diagrams do not contain loops that involve type-(ii) vertices only.

Typical diagrams are presented in Fig. 2.

IV. EFFECTIVE ACTION OF A MANY-FERMION SYSTEM BASED ON THE REDUCED SINGLE-ELECTRON DENSITY MATRIX

In this section we apply the effective action approach of Sec. II to a general many-fermion system. Consider a non-relativistic system of M electrons that can occupy N possible states ($M \leq N$) denoted with latin indices with overbars (\bar{n}, \bar{m}, \dots). The states are assumed to be orthonormal and the indices include the spin. We can recast the total Hamiltonian as

$$\hat{H}_T(\tau) = \sum_{\bar{m}, \bar{n}} t_{\bar{m}\bar{n}} \hat{c}_{\bar{m}}^\dagger \hat{c}_{\bar{n}} + \sum_{\bar{m}, \bar{n}, \bar{k}, \bar{l}} \bar{V}_{\bar{m}\bar{n}\bar{k}\bar{l}} \hat{c}_{\bar{m}}^\dagger \hat{c}_{\bar{n}}^\dagger \hat{c}_{\bar{k}} \hat{c}_{\bar{l}} - \sum_{\bar{m}, \bar{n}} \mathcal{E}_{\bar{m}\bar{n}}(\tau) \hat{c}_{\bar{m}}^\dagger \hat{c}_{\bar{n}}, \quad (4.1a)$$

where the electron annihilation (creation) operators \hat{c} (\hat{c}^\dagger) satisfy the commutation relations of fermions

$$\hat{c}_{\bar{m}} \hat{c}_{\bar{n}}^\dagger + \hat{c}_{\bar{n}}^\dagger \hat{c}_{\bar{m}} = \delta_{\bar{m}\bar{n}}; \quad (4.1b)$$

other anticommutators of \hat{c} and \hat{c}^\dagger are zero. The matrix $\bar{V}_{\bar{m}\bar{n}\bar{k}\bar{l}}$ is taken to be antisymmetric with respect to permutations \bar{m} with \bar{n} and \bar{k} with \bar{l} .

We adopt the multipolar form of the total Hamiltonian describing the material system and quantum transverse electromagnetic field, neglecting magnetic terms [2]. The multipolar Hamiltonian can be written in terms of the position-dependent polarization $\hat{P}(\mathbf{r})$, which will be defined for the system of Eqs. (4.1) $\hat{P}(\mathbf{r}) = \sum_{\bar{m}, \bar{n}} \mu_{\bar{m}\bar{n}}(\mathbf{r}) \hat{c}_{\bar{m}}^\dagger \hat{c}_{\bar{n}}$, where $\mu_{\bar{m}\bar{n}}(\mathbf{r}) = \int_0^1 d\lambda \int d\mathbf{r}' \mathbf{r}' \delta(\mathbf{r} - \lambda \mathbf{r}') \rho_{\bar{m}\bar{n}}(\mathbf{r}')$ and $\rho_{mn}(\mathbf{r}')$ are matrix elements of the charge-density operator $\hat{\rho}(\mathbf{r}')$, which can be expanded in the form $\hat{\rho}(\mathbf{r}') = \sum_{\bar{m}, \bar{n}} \rho_{\bar{m}\bar{n}}(\mathbf{r}') \hat{c}_{\bar{m}}^\dagger \hat{c}_{\bar{n}}$. The interaction with the external transverse field given by $-\int d\mathbf{r} \mathcal{E}(\mathbf{r}, t) \cdot \hat{P}(\mathbf{r})$ then yields the last term in Eq. (4.1a), where $\mathcal{E}_{\bar{m}\bar{n}}(\tau) \equiv \int d\mathbf{r} \mathcal{E}(\mathbf{r}, \tau) \mu_{\bar{m}\bar{n}}(\mathbf{r})$.

The effective action formalism starts with a choice of a set of operators \hat{X} whose expectation values x form the variables of the effective action. It is natural to choose these operators to be related to the complete set of variables of the classical analog of the quantum system. To apply this to the system of Eqs. (4.1), we will make use of the fact that the time-dependent Hartree-Fock equations are the dynamical equations describing the classical analog of our quantum system. This allows us to treat the corrections to the TDHF approach as a "semiclassical" expansion of the effective action. The TDHF equations are equations of motion for the

reduced electron density matrix $\rho_{\bar{m}\bar{n}}$, which has the form of a projector onto an M -dimensional subspace $V(\rho)$ of the N -dimensional space of possible single-electron states, and can be associated with a single Slater determinant $\Omega(\rho)$ [12]. In other words, $\rho_{\bar{m}\bar{n}}$ is an $N \times N$ Hermitian matrix with rank M and $\rho^2 = \rho$, and belongs to the phase space of a classical dynamical system that forms a Grassman manifold $G(M, N; C)$, defined as the set of all M -dimensional complex vector subspaces of a complex N -dimensional vector space [41]. The quantum system of Eqs. (4.1) can be obtained from the above-mentioned classical system by applying the procedure of geometrical quantization [41]. In particular, we can use the path-integral representation of Sec. II, assuming that q belongs to the Grassman manifold.

Since the Grassman manifold has a nontrivial geometrical structure, we need to introduce a system of local coordinates. To that end we first choose the origin of the coordinate system $\rho^{(0)}$, which is a point in the Grassman manifold. We then define a basis set $e_{\bar{m}}$, $\bar{m} = 1, \dots, N$, in the space of single-electron states so that e_{m_2} , $m_2 = 1, \dots, M$, belong to $V(\rho^{(0)})$ and e_{m_1} , $m_1 = M+1, \dots, N$, are orthogonal to $V(\rho^{(0)})$. Hereafter, we use the following convention: latin indices with overbars assume the values $\bar{m} = 1, \dots, N$ and latin indices with subscripts 1 and 2 assume the values $m_1 = M+1, \dots, N$ and $m_2 = 1, \dots, M$. In this notation $\rho_{m_1 m_2}^{(0)} = \rho_{m_2 m_1}^{(0)} = 0$. We define the coordinates of the reduced density matrix ρ as its particle-hole components $\rho_{m_1 m_2}$ and $\rho_{m_2 m_1} = \rho_{m_1 m_2}^*$. The number of complex coordinates is $M(N-M)$, which coincides with the complex dimension of the Grassman manifold $G(M, N; C)$.

At this point we introduce a new set of latin subscripts that represent pairs of indices denoting the particle-hole modes

$$m \equiv (m_1, m_2), \quad m_1 = M+1, \dots, N, \quad m_2 = 1, \dots, M. \quad (4.2)$$

To be consistent with the notation of Sec. II we define

$$X_m = \rho_{m_2 m_1}, \quad \bar{X}_m = X_m^* = \rho_{m_1 m_2}. \quad (4.3a)$$

Since the classical variables X_m in the geometric quantization scheme correspond to the operators $\hat{c}_{m_1}^\dagger \hat{c}_{m_2}$, we define the set of \hat{X} operators as

$$\hat{X}_m \equiv \hat{c}_{m_2}^\dagger \hat{c}_{m_1}, \quad \hat{\bar{X}}_m \equiv \hat{X}_m^\dagger = \hat{c}_{m_1}^\dagger \hat{c}_{m_2}. \quad (4.3b)$$

Once the set of operators \hat{X} is defined, we can follow the procedure of Sec. II and define the effective action $S_{\text{eff}}[x_{a\pm}(\tau), \bar{x}_{a\pm}(\tau); \rho^{(0)}]$, which depends on $\rho^{(0)}$ parametrically. Equations (2.11) then become

$$\frac{\delta S_{\text{eff}}}{\delta x_{a-}(\tau)} = \bar{h}_{a+}(\tau), \quad \frac{\delta S_{\text{eff}}}{\delta \bar{x}_{a-}(\tau)} = h_{a+}(\tau), \quad (4.4a)$$

$$\frac{\delta S_{\text{eff}}}{\delta x_{a+}(\tau)} = \bar{h}_{a-}(\tau), \quad \frac{\delta S_{\text{eff}}}{\delta \bar{x}_{a+}(\tau)} = h_{a-}(\tau). \quad (4.4b)$$

At this point we need to determine $\rho^{(0)}$. It follows from the way the effective action is constructed that $\rho^{(0)}$ defines the effective action variables. This means that the parametric dependence of S_{eff} on $\rho^{(0)}$ reflects the dependence of S_{eff} on the choice of variables. The optical response is related to deviations of the reduced density matrix from its ground-state form $\rho^{(g)}$, induced by an external field. It is therefore natural to relate the choice of variables to the ground-state properties so that $x_a(\tau), \bar{x}_a(\tau)$ represent deviations from the ground-state values, i.e., in the absence of the external field $x_{a\pm}(t) = \bar{x}_{a\pm}(t) \equiv 0$. This implies $\rho_{m_1 m_2}^{(g)} = \rho_{m_2 m_1}^{(g)} = 0$. Utilizing the fact that $\rho^{(0)}$ is a projection operator, we obtain the following procedure for expressing $\rho^{(0)}$ in terms of $\rho^{(g)}$. Let l_1, l_2, \dots, l_n be a basis set in the space of single-electron states in which $\rho^{(g)}$ is diagonal, i.e., $\rho^{(g)} l_{\bar{m}} = \lambda_{\bar{m}} l_{\bar{m}}$. This is known as the natural orbitals basis [37–39]. Note that $0 \leq \lambda_{\bar{m}} \leq 1$ for all \bar{m} and $\sum_{\bar{m}=1} \lambda_{\bar{m}} = M$. We assume that the basis functions are ordered as follows: $\lambda_{\bar{m}} < \lambda_{\bar{n}}$ for $\bar{m} > \bar{n}$. We then define an M -dimensional vector subspace $V^{(0)}$ generated by vectors l_1, l_2, \dots, l_M . $\rho^{(0)}$ is the projector operator on $V^{(0)}$ and we have $V(\rho^{(0)}) = V^{(0)}$. Stated differently, $\rho^{(0)}$ is the projector into the subspace generated by the mostly occupied natural orbitals. This means that we can immediately find $\rho^{(0)}$ once $\rho^{(g)}$ is given and the variables of S_{eff} are defined with respect to the ground state reduced density matrix. We thus obtain a unique prescription for determining the effective action from $\rho^{(g)}$. It follows from Eqs. (4.4) for the zero external field that

$$\left. \frac{\delta S_{\text{eff}}(\rho^{(0)})}{\delta x_{a-}(\tau)} \right|_{x_{\pm} = \bar{x}_{\pm} = 0} = 0, \quad \left. \frac{\delta S_{\text{eff}}(\rho^{(0)})}{\delta \bar{x}_{a-}(\tau)} \right|_{x_{\pm} = \bar{x}_{\pm} = 0} = 0. \quad (4.5)$$

Equations (4.5) imply that the linear terms in the expansion of S_{eff} in powers of x, \bar{x} vanish and that the leading terms are quadratic.

As indicated in Sec. II, Eqs. (4.4) enable us to calculate the optical response of our quantum system once the effective action is known. If we are interested in the response to a field coupled to particle-hole components only, we can use the simpler system of equations that follows from Eq. (2.12):

$$\left. \frac{\delta S_{\text{eff}}}{\delta x_{a-}(\tau)} \right|_{x_{\pm} = \bar{x}_{\pm} = 0} = \bar{h}_a(\tau), \quad \left. \frac{\delta S_{\text{eff}}}{\delta \bar{x}_{a-}(\tau)} \right|_{x_{\pm} = \bar{x}_{\pm} = 0} = h_a(\tau). \quad (4.6)$$

Finally, we emphasize that in this section we assumed that $\rho^{(g)}$ is known. We can then use any of the standard (e.g., *ab initio* or density functional) methods for computing the ground-state reduced density matrix as an input to our calculation of the response. However, we will show in Sec. VIII that the effective action approach also allows us to develop a self-consistent procedure for calculating ρ_g .

V. PERTURBATIVE EXPANSION OF THE EFFECTIVE ACTION: ELECTRON-HOLE CONTRIBUTIONS TO THE LINEAR RESPONSE

In this section we derive expressions for the linear response using a perturbative calculation of the effective action in powers of x and \bar{x} . Equations (4.5) imply that the linear

terms vanish and to obtain the linear response with respect to a field coupled to the particle-hole components we need only the quadratic terms. A systematic procedure for calculating the effective action is obtained by partitioning the Hamiltonian \hat{H} [Eq. (4.1)] in the form of Eq. (3.1), where \hat{H}_0 contains the terms that conserve the number of electron-hole pairs [with respect to the single Slater determinant $\Omega(\rho^{(0)})$] and \hat{H}_1 contains the remaining terms. The effective action can be calculated perturbatively in \hat{H}_1 , as outlined in Sec. III. The quadratic terms of the expansion of S_{eff} will be calculated to first nonvanishing order, in Appendix A. The particle-particle and hole-hole contributions to the linear response show up only in higher orders and will be calculated in Sec. VII. Therefore, we only need the quadratic terms in the expansion of S_{eff} and may work with the simpler equations [Eq. (4.6)]. The quadratic part of the effective action has the form $S_{\text{eff}} = S_I + S_{II}$, where

$$\begin{aligned} S_I = & i \int \bar{x}_{a-}(\tau) \frac{\partial x_{a+}(\tau)}{\partial \tau} d\tau - i \int x_{a-}(\tau) \frac{\partial \bar{x}_{a+}(\tau)}{\partial \tau} d\tau \\ & - \int d\tau L_{ab} \bar{x}_{a-}(\tau) x_{b+}(\tau) - \int d\tau L_{ab}^* x_{a-}(\tau) \bar{x}_{b+}(\tau) \\ & - 2 \int d\tau \Delta_{ab}^{(0)} \bar{x}_{a+}(\tau) \bar{x}_{b-}(\tau) \\ & - 2 \int d\tau (\Delta_{ab}^{(0)})^* x_{a+}(\tau) x_{b-}(\tau) \end{aligned} \quad (5.1a)$$

and

$$\begin{aligned} S_{II} = & - \int d\tau'' d\tau' \bar{x}_{a-}(\tau'') x_{b+}(\tau') \Sigma_{ab}(\tau'' - \tau') \\ & - \int d\tau'' d\tau' \bar{x}_{a+}(\tau'') x_{b-}(\tau') \Sigma_{ba}^*(\tau' - \tau'') \\ & - 2 \int d\tau \bar{\Delta}_{ab} \bar{x}_{a+}(\tau) \bar{x}_{b-}(\tau) \\ & - 2 \int d\tau \bar{\Delta}_{ab}^* x_{a+}(\tau) x_{b-}(\tau) + S_i. \end{aligned} \quad (5.1b)$$

Expressions for $\Delta^{(0)}$, $\bar{\Delta}$, L , and Σ are presented below. Hereafter, we adopt a convention of summation over repeating indices. S_i contains the terms with x_- and \bar{x}_- components only, which do not contribute to Eqs. (4.6). Substituting Eqs. (5.1) into Eqs. (4.6), we obtain equations of motion that determine the linear optical response

$$\begin{aligned} i \frac{\partial x_a(\tau)}{\partial \tau} - L_{ab} x_b(\tau) - \int d\tau' \Sigma_{ab}(\tau - \tau') x_b(\tau') \\ - 2 \Delta_{ab} \bar{x}_b(\tau) = h_a(\tau), \end{aligned} \quad (5.2a)$$

$$\begin{aligned} -i \frac{\partial \bar{x}_a(\tau)}{\partial \tau} - L_{ab}^* \bar{x}_b(\tau) - \int d\tau' \Sigma_{ab}^*(\tau - \tau') \bar{x}_b(\tau') \\ - 2 \Delta_{ab}^* \bar{x}_b(\tau) = \bar{h}_a(\tau), \end{aligned} \quad (5.2b)$$

where

$$\Delta_{ab} \equiv \Delta_{ab}^{(0)} + \bar{\Delta}_{ab}. \quad (5.3)$$

Introducing the ‘‘momentum-coordinate’’ variables

$$P_a \equiv -\frac{i}{\sqrt{2}}(x_a - \bar{x}_a), \quad (5.4a)$$

$$Q_a \equiv \frac{1}{\sqrt{2}}(x_a + \bar{x}_a)$$

and the corresponding components of the external field

$$\varepsilon_a(\tau) = \frac{1}{\sqrt{2}}[h_a(\tau) + \bar{h}_a(\tau)], \quad (5.4b)$$

$$\tilde{\varepsilon}_a(\tau) = -\frac{i}{\sqrt{2}}[h_a(\tau) - \bar{h}_a(\tau)],$$

we can recast Eqs. (5.2) in the form of coupled classical oscillators

$$\begin{aligned} \frac{\partial Q_a(\tau)}{\partial \tau} - \text{Re}(L_{ab}) P_b(\tau) - \int d\tau' \text{Re} \Sigma_{ab}(\tau - \tau') P_b(\tau') \\ - 2 \text{Im}(\Delta_{ab}) Q_b(\tau) - \text{Im}(L_{ab}) Q_b(\tau) \\ - \int d\tau' \text{Im} \Sigma_{ab}(\tau - \tau') Q_b(\tau') \\ + 2 \text{Re}(\Delta_{ab}) P_b(\tau) = \tilde{\varepsilon}_a(\tau), \end{aligned} \quad (5.5a)$$

$$\begin{aligned} \frac{\partial P_a(\tau)}{\partial \tau} + \text{Re}(L_{ab}) Q_b(\tau) + \int d\tau' \text{Re} \Sigma_{ab}(\tau - \tau') Q_b(\tau') \\ + 2 \text{Im}(\Delta_{ab}) P_b(\tau) - \text{Im}(L_{ab}) P_b(\tau) \\ - \int d\tau' \text{Im} \Sigma_{ab}(\tau - \tau') P_b(\tau') \\ + 2 \text{Re}(\Delta_{ab}) Q_b(\tau) = -\varepsilon_a(\tau). \end{aligned} \quad (5.5b)$$

The linear response is obtained by solving Eqs. (5.2) after switching to the frequency domain. Assuming $\bar{h}_a = h_a$ (which implies that the external field interacts with oscillator coordinates, but not momenta), we have $\mu_a^*(\mathbf{r}) = \mu_a(\mathbf{r})$ [i.e., $\mu_a(\mathbf{r})$ is real] and obtain

$$\chi^{(1)}(\mathbf{r}, \mathbf{r}'; \omega) = \sum_{a,b} \mu_a(\mathbf{r}) \mu_b(\mathbf{r}') \chi_{ab}(\omega), \quad (5.6a)$$

where

$$\mu_a(\mathbf{r}) \equiv \mu_{a_1 a_2}(\mathbf{r}) \quad (5.6b)$$

and

$$\chi(\omega) = \chi^{(11)}(\omega) + \chi^{(12)}(\omega) + \chi^{(21)}(\omega) + \chi^{(22)}(\omega), \quad (5.7a)$$

where

$$\begin{pmatrix} \chi^{(11)}(\omega) & \chi^{(12)}(\omega) \\ \chi^{(21)}(\omega) & \chi^{(22)}(\omega) \end{pmatrix} = \begin{pmatrix} \omega - L - \Sigma(\omega) & -2\Delta \\ -2\Delta^\dagger & -\omega - L^* - \Sigma^*(-\omega) \end{pmatrix}^{-1}. \quad (5.7b)$$

Here $\chi^{(ij)}$, L , $\Sigma(\omega)$, and Δ are matrices with $L_{mn} = L_{nm}^*$, $\Sigma_{mn}(\omega) = \Sigma_{nm}^*(\omega)$, $\Delta_{mn} = \Delta_{nm}$, $(\Delta^\dagger)_{mn} = \Delta_{nm}^*$, and $(L^*)_{mn} = L_{mn}^*$. $\bar{\Sigma}(\omega)$ is the Fourier transform of $\Sigma(\tau - \tau')$

$$\bar{\Sigma}_{ab}(\tau - \tau') \equiv \int \frac{d\omega}{2\pi} \exp[-i\omega(\tau - \tau')] \Sigma_{ab}(\omega). \quad (5.7c)$$

The quantities L , Σ , $\Delta^{(0)}$, and $\bar{\Delta}$ are given by

$$\begin{aligned} L_{mn} = & \bar{t}_{m_1 n_1} \delta_{m_2 n_2} - \bar{t}_{m_2 n_2} \delta_{m_1 n_1} - 2\bar{V}_{m_1 k_2 n_1 k_2} \delta_{m_2 n_2} \\ & + 2\bar{V}_{m_1 n_2 n_1 m_2} + 2\bar{V}_{n_2 k_2 m_2 k_2} \delta_{m_1 n_1}, \end{aligned} \quad (5.8)$$

$$\Delta_{ab}^{(0)} = (I - P)_{aba'b'} V_{a'b'}, \quad (5.9a)$$

$$\bar{\Delta}_{ab} = 2\bar{\Gamma}_{abmn}^{(b)}(0) \bar{F}_{mnm'n'}(0) (I - P)_{m'n'a'b'} V_{a'b'}, \quad (5.9b)$$

and

$$\bar{\Sigma}_{ab}(\omega) = \Sigma_{ab}^{(1)}(\omega) + \Sigma_{ab}^{(2)}(\omega), \quad (5.10a)$$

with

$$\begin{aligned} \Sigma_{ab}^{(1)}(\omega) = & W_{mna}^* W_{m'n'b} \{ 2\bar{F}_{mnm'n'}(\omega) + 4\bar{F}_{mnkl}(\omega) \\ & \times \bar{\Gamma}_{k'ik'l'}^{(b)}(\omega) \bar{F}_{k'l'm'n'}(\omega) \}, \end{aligned} \quad (5.10b)$$

$$\begin{aligned} \Sigma_{ab}^{(2)}(\omega) = & -16\Delta_{pq} \Delta_{cd}^* \int \frac{d\omega'}{2\pi i} G_{m'p}(-\omega') G_{cq}(\omega') \\ & \times G_{dn'}(-\omega') \bar{\Gamma}_{an'bm'}^{(s)}(\omega - \omega'). \end{aligned} \quad (5.10c)$$

In Eqs. (5.9) and (5.10) I is a unit matrix and P is the electron permutation matrix

$$P_{aba'b'} \equiv \delta_{a_1 b_1'} \delta_{b_1 a_1'} \delta_{a_2 a_2'} \delta_{b_2 b_2'}. \quad (5.11)$$

The single-mode Green function $G(\omega)$ is given by

$$G_{mn}(\omega) = [(\omega - L + i\eta)^{-1}]_{mn}, \quad (5.12)$$

and the two-mode bare Green function $\bar{F}(\omega)$ is defined by

$$\bar{F}_{mnm'n'}(\omega) = \frac{1}{2} [\bar{F}_{mnm'n'}^{(0)}(\omega) + \bar{F}_{nmm'n'}^{(0)}(\omega)], \quad (5.13a)$$

$$\bar{F}_{mnm'n'}^{(0)}(\omega) = - \int \frac{d\omega'}{2\pi i} G_{mn'}(\omega') G_{nn'}(\omega - \omega'). \quad (5.13b)$$

The scattering matrix of two modes $\bar{\Gamma}_{mnm'n'}^{(b)}(\omega)$ has the operator form

$$\bar{\Gamma}^{(b)}(\omega) = U[I - 2\bar{F}(\omega)U]^{-1}. \quad (5.14)$$

The scattering matrix of two excitations $\bar{\Gamma}_{mnm'n'}^{(s)}(\omega)$ can be represented in two equivalent forms

$$\bar{\Gamma}^{(s)}(\omega) = -\frac{1}{2} P[\bar{F}(\omega)]^{-1} + \bar{\Gamma}^{(b)}(\omega) \bar{F}(\omega) (I - P)[\bar{F}(\omega)]^{-1}, \quad (5.15a)$$

$$\bar{\Gamma}^{(s)}(\omega) = -\frac{1}{2} [\bar{F}(\omega)]^{-1} P + [\bar{F}(\omega)]^{-1} (I - P) \bar{F}(\omega) \bar{\Gamma}^{(b)}(\omega). \quad (5.15b)$$

The matrices U , W , and V are expressed in terms of the Hamiltonian parameters in the following way:

$$U_{mnm'n'} = \frac{1}{4} (U_{mnm'n'}^{(0)} + U_{nmn'm'}^{(0)} + U_{mnn'm'}^{(0)} + U_{nmm'n'}^{(0)}), \quad (5.16a)$$

$$\begin{aligned} U_{mnm'n'}^{(0)} = & 2\bar{V}_{m_1 n_2' m_1' n_2} \delta_{m_2 m_2'} \delta_{n_1 n_1'} - 2\bar{V}_{m_1 n_1 m_1' n_1'} \delta_{m_2 m_2'} \delta_{n_2 n_2'} \\ & - 2\bar{V}_{m_2' n_2' m_2 n_2} \delta_{m_1 m_1'} \delta_{n_1 n_1'}, \end{aligned} \quad (5.16b)$$

$$W_{mna} = \frac{1}{2} (W_{mna}^{(0)} + W_{nma}^{(0)}), \quad (5.17a)$$

$$\begin{aligned} W_{mna}^{(0)} = & \frac{1}{\sqrt{2}} \bar{V}_{m_1 n_1 a_1 m_2} \delta_{n_2 a_2} - \frac{1}{\sqrt{2}} \bar{V}_{m_1 n_1 a_1 n_2} \delta_{m_2 a_2} \\ & - \frac{1}{\sqrt{2}} \bar{V}_{a_2 m_1 m_2 n_2} \delta_{n_1 a_1} + \frac{1}{\sqrt{2}} \bar{V}_{a_2 m_1 n_2 m_2} \delta_{n_1 a_1}, \end{aligned} \quad (5.17b)$$

and

$$V_{mn} = \frac{1}{2} \bar{V}_{m_1 n_1 n_2 m_2} \frac{1}{2} \bar{V}_{n_1 m_1 m_2 n_2}. \quad (5.18)$$

Equations (5.6) and (5.7), together with Eqs. (5.8)–(5.18), define the electron-hole contribution to the linear response of the system. The TDHF procedure is obtained by retaining the action S_I and neglecting S_{II} . This implies setting $\Sigma(\omega) = 0$ and $\Delta = \Delta^{(0)}$ in Eq. (5.7b). These expressions can be simplified using the basis set of eigenmodes of L in the space of particle-hole modes, where

$$L_{mn} = \delta_{mn} \omega_n, \quad (5.19a)$$

$$G_{mn}(\omega) = \frac{\delta_{mn}}{\omega - \omega_n + i\eta}, \quad (5.19b)$$

and

$$\begin{aligned} \bar{F}_{mnm'n'}(\omega) = & \frac{1}{2} (\delta_{mm'} \delta_{nn'} + \delta_{mn'} \delta_{nm'}) \\ & \times \frac{1}{\omega - (\omega_m + \omega_n) + i\eta}, \end{aligned} \quad (5.20a)$$

$$\begin{aligned} \Sigma_{ab}^{(2)}(\omega) = & 16 \sum_{p,q,l} \Delta_{pl} \Delta_{lq}^* \bar{\Gamma}_{qabp}^{(s)}(\omega - \omega_l) \\ & \times \frac{1}{\omega_p + \omega_l} \frac{1}{\omega_q + \omega_l}. \end{aligned} \quad (5.20b)$$

VI. EIGENMODE EXPANSION OF THE LINEAR RESPONSE

We now express the linear response in terms of eigenvalues and eigenmodes of the linear problem. In this form the linear response is represented as a sum of resonant terms, which is reminiscent of sum over eigenstates of the system

[6]. To that end we note that the linear response is expressed in terms of a matrix $A^{-1}(\omega)$, where

$$A(\omega) \equiv \begin{pmatrix} \omega - L - \Sigma(\omega) & -2\Delta \\ -2\Delta^\dagger & -\omega - L^* - \Sigma^*(-\omega) \end{pmatrix} \quad (6.1)$$

[see Eqs. (5.7)]. The frequency-dependent matrix $A(\omega)$ acts in the space of vectors ξ with components ξ_{1a}, ξ_{2a} . The matrix is Hermitian for real ω with respect to the scalar product

$$(\xi, \eta) \equiv \eta_{1a}^* \xi_{1a} + \eta_{2a}^* \xi_{2a}. \quad (6.2)$$

Since $A(\omega)$ is a meromorphic matrix function of ω , $A^{-1}(\omega)$ is meromorphic too, and can be expanded in the form

$$A^{-1}(\omega) = \sum_{\alpha} \frac{B_{\alpha}}{\omega - \omega_{\alpha} + i\eta}, \quad (6.3)$$

where B_{α} are matrices, η stands for small damping rate, and the discrete set of frequencies $\{\omega_{\alpha}\}$ is the spectrum of $A(\omega)$, i.e., the values of ω for which $A(\omega)$ cannot be inverted. This means that for each ω_{α} , one can find a vector (eigenmode) ξ_{α} for which

$$A(\omega_{\alpha})\xi_{\alpha} = 0. \quad (6.4a)$$

Equation (6.4a) has the following form, once the components are specified:

$$[\omega_{\alpha} \delta_{ab} - L_{ab} - \Sigma_{ab}(\omega_{\alpha})]\xi_{1b\alpha} - 2\Delta_{ab}\xi_{2b\alpha} = 0, \quad (6.4b)$$

$$-2\Delta_{ab}^*\xi_{1b\alpha} + [-\omega_{\alpha} \delta_{ab} - L_{ab}^* - \Sigma_{ab}^*(-\omega_{\alpha})]\xi_{2b\alpha} = 0. \quad (6.4c)$$

Considering $A(\omega)$ near an eigenvalue ω_{α} and making use of the representation Eq. (6.3), we obtain

$$B_{\alpha} = \frac{|\xi_{\alpha}\rangle\langle\xi_{\alpha}|}{\left\langle \xi_{\alpha} \left| \frac{dA(\omega)}{d\omega} \right|_{\omega=\omega_{\alpha}} \right| \xi_{\alpha} \rangle}. \quad (6.5)$$

From Eqs. (6.3) and (6.5) and using the fact that for each eigenmode ξ_{α} the vector $\xi_{-\alpha}$ with components $\xi_{1a, -\alpha} \equiv \xi_{2a, \alpha}^*$ and $\xi_{2a, -\alpha} \equiv \xi_{1a, \alpha}^*$ is an eigenmode with $\omega_{-\alpha} \equiv -\omega_{\alpha}$, we recast the linear response $\chi(\omega)$ in the form

$$\chi(\mathbf{r}, \mathbf{r}'; \omega) = \sum_{\alpha > 0} \left\{ \frac{\mu_{\alpha}(\mathbf{r})\mu_{\alpha}^*(\mathbf{r}')}{\omega - \omega_{\alpha} + i\eta} - \frac{\mu_{\alpha}^*(\mathbf{r})\mu_{\alpha}(\mathbf{r}')}{\omega + \omega_{\alpha} + i\eta} \right\}, \quad (6.6)$$

where $\sum_{\alpha > 0}$ implies summation over eigenmodes with $\omega_{\alpha} > 0$. $\mu_{\alpha}(\mathbf{r})$ has the form

$$\mu_{\alpha}(\mathbf{r}) \equiv \xi_{1a}\mu_a(\mathbf{r}) + \xi_{2a}\mu_a(\mathbf{r}) \quad (6.7a)$$

and the modes ξ_{α} should be normalized using the condition

$$\xi_{1a\alpha}^* \xi_{1a\alpha} - \xi_{2a\alpha}^* \xi_{2a\alpha} - \frac{\partial \Sigma_{ab}(\omega)}{\partial \omega} \Big|_{\omega=\omega_{\alpha}} \xi_{1a\alpha}^* \xi_{1b\alpha} - \frac{\partial \Sigma_{ab}^*(-\omega)}{\partial \omega} \Big|_{\omega=\omega_{\alpha}} \xi_{2a\alpha}^* \xi_{2b\alpha} = 1. \quad (6.7b)$$

Equation (6.7b) contains a summation over repeating latin indices but no summation over the greek indices α .

VII. PARTICLE-PARTICLE AND HOLE-HOLE CONTRIBUTIONS TO THE LINEAR RESPONSE

In Secs. V and VI we obtained expressions for the linear optical response, assuming that the external field is coupled only to the particle-hole components of the reduced density matrix. Coupling to the particle-particle and hole-hole components leads to higher-order contributions in the parameters W and V . These contributions are of great interest since they vanish within the TDHF approximation and can induce new resonances by introducing oscillator strength into resonances that are dark in the TDHF approximation.

In this section we present the linear response, taking into account coupling to all components of the reduced density matrix $\hat{X}_{\bar{m}\bar{n}}$ and calculating the corrections to lowest order in the parameters W and V . Making use of the correlation function expressions for the optical response [Eqs. (2.4)] and the expression for the polarization that follows from Eq. (4.1a), we can write the linear response in the form

$$\chi(\mathbf{r}, \mathbf{r}'; \omega) = -i \sum_{\bar{m}, \bar{n}, \bar{m}', \bar{n}'} \int_0^{\infty} d\tau \mu_{\bar{m}\bar{n}}(\mathbf{r}) \mu_{\bar{m}'\bar{n}'}(\mathbf{r}') \times \langle \hat{X}_{\bar{m}\bar{n}+}(\tau) \hat{X}_{\bar{m}'\bar{n}'-}(0) \rangle \exp(i\omega\tau). \quad (7.1)$$

The \hat{X} operators have particle-particle components [operators \hat{X}_m and \hat{X}_m defined by Eqs. (4.3b)] as well as particle-particle or hole-hole components (operators $\hat{X}_{a_j b_j} \equiv \hat{c}_{a_j}^\dagger \hat{c}_{b_j}$, $J=1,2$). The correlation functions on the rhs of Eq. (7.1) for different components of \hat{X} are calculated in Sec. V and Appendix C. Substituting the correlation functions into Eq. (7.1), we obtain the linear response. This expression contains a large number of terms; however, by making some transformations, it can be written in a compact matrix form, which is most suitable for the eigenmode expansion of Sec. VI. We start by introducing a frequency-dependent matrix $B(\omega)$ acting on the space \mathscr{W} , which can be decomposed in the following way:

$$\mathscr{W} = \mathscr{W}_1 \oplus \bar{\mathscr{W}}_1 \oplus \mathscr{W}_2 \oplus \bar{\mathscr{W}}_2, \quad (7.2)$$

where $\mathscr{W}_1 \cong \bar{\mathscr{W}}_1$ are two replicas of the single mode space

and $\mathcal{W}_2 \equiv \bar{\mathcal{W}}_2 \equiv \mathcal{W}_1 \otimes \mathcal{W}_1$ are two replicas of the two-mode space. Equation (7.2) leads to the canonical decomposition of the operator $B(\omega): \mathcal{W} \rightarrow \mathcal{W}$

$$B(\omega) = \sum_{U, V \in A} B_{UV}(\omega), \quad (7.3)$$

where A is a set $A = \{\mathcal{W}_1, \bar{\mathcal{W}}_1, \mathcal{W}_2, \bar{\mathcal{W}}_2\}$ and $B_{UV}: V \rightarrow U$. The operator $B(\omega)$ is Hermitian for real ω , which means

$$[B_{UV}(\omega)]^\dagger = B_{VU}(\omega). \quad (7.4)$$

We will denote with ξ_{1a} , ξ_{2a} , ξ_{1mn} , and ξ_{2mn} the components of a vector $\xi \in \mathcal{W}$ related to the subspaces \mathcal{W}_1 , $\bar{\mathcal{W}}_1$, \mathcal{W}_2 , and $\bar{\mathcal{W}}_2$, respectively. The scalar product on \mathcal{W} is the sum of the products of the components.

The nonzero components of $B(\omega)$ are defined as

$$\begin{aligned} B_{\mathcal{W}_1 \mathcal{W}_1}(\omega) &= \omega - L - \Sigma^{(2)}(\omega), \\ B_{\bar{\mathcal{W}}_1 \bar{\mathcal{W}}_1}(\omega) &= -\omega - L^* - \Sigma^{(2)*}(-\omega); \end{aligned} \quad (7.5a)$$

$$B_{\mathcal{W}_2 \mathcal{W}_2}(\omega) = \omega - \bar{L}, \quad B_{\bar{\mathcal{W}}_2 \bar{\mathcal{W}}_2}(\omega) = -\omega - \bar{L}^*; \quad (7.5b)$$

$$B_{\mathcal{W}_1 \bar{\mathcal{W}}_1}(\omega) = -2\Delta, \quad B_{\bar{\mathcal{W}}_1 \mathcal{W}_1}(\omega) = -2\Delta^\dagger; \quad (7.5c)$$

$$B_{\mathcal{W}_2 \bar{\mathcal{W}}_1}(\omega) = -W, \quad B_{\bar{\mathcal{W}}_2 \mathcal{W}_1}(\omega) = -W^*; \quad (7.5d)$$

$$B_{\mathcal{W}_1 \mathcal{W}_2}(\omega) = -W^\dagger, \quad B_{\bar{\mathcal{W}}_1 \bar{\mathcal{W}}_2}(\omega) = -(W^*)^\dagger, \quad (7.5e)$$

where the operators L and Δ are defined in Sec. V and W has the matrix elements $W_{mn,a} \equiv W_{mna}$. The operator \bar{L} is defined as the projection of the operator \bar{H}_0 into the subspace of two-mode states. The Green function $\bar{G}_{mnm'n'}(\omega)$ of \bar{L} is

$$\bar{G}_{mnm'n'}(\omega) \equiv [(\omega - \bar{L} + i\eta)^{-1}]_{mnm'n'}, \quad (7.6a)$$

which has the form

$$\begin{aligned} \bar{G}_{mnm'n'}(\omega) &= 2\bar{F}_{mnm'n'}(\omega) + 4\bar{F}_{mnpq}(\omega) \\ &\quad \times \bar{\Gamma}_{pp'q'q'}^{(b)}(\omega) \bar{F}_{p'q'm'n'}(\omega). \end{aligned} \quad (7.6b)$$

Equations (7.5) can be written in matrix form

$$B(\omega) = \begin{pmatrix} \omega - \bar{L} & -W & 0 & 0 \\ W^\dagger & \omega - L - \Sigma^{(2)}(\omega) & -2\Delta & 0 \\ 0 & -2\Delta^\dagger & -\omega - L^* - \Sigma^{(2)*}(-\omega) & (W^*)^\dagger \\ 0 & 0 & W^* & -\omega - \bar{L}^* \end{pmatrix}. \quad (7.7)$$

Let $\chi(\omega)$ be the inverse of the $B(\omega)$ operator

$$\chi(\omega) \equiv B^{-1}(\omega), \quad (7.8)$$

with the components $\chi_{m,n}^{(ij)}(\omega)$, $\chi_{mn,a}^{(ij)}(\omega)$, $\chi_{a,mn}^{(ij)}(\omega)$, and $\chi_{mn,m'n'}^{(ij)}(\omega)$ with $i, j = 1, 2$. The linear optical response function $\chi(\mathbf{r}, \mathbf{r}'; \omega)$ can then be expressed in terms of the operator $\chi(\omega)$

$$\chi(\mathbf{r}, \mathbf{r}'; \omega) = \text{Tr}[\mu^\dagger(\mathbf{r}) \chi(\omega) \mu(\mathbf{r}')], \quad (7.9a)$$

with $\mu_a^\dagger(\mathbf{r}) \equiv \mu_a^*(\mathbf{r})$ and $\mu_{mn}^\dagger(\mathbf{r}) \equiv \mu_{nm}^*(\mathbf{r})$. $\chi(\omega)$ is given by Eq. (7.8) and the components of $\mu(\mathbf{r})$ are

$$\mu_{1a}(\mathbf{r}) = \mu_{2a}(\mathbf{r}) \equiv \mu_a(\mathbf{r}), \quad (7.9b)$$

$$\begin{aligned} \mu_{1ab}(\mathbf{r}) &= (\mathcal{L}_{m_1 n_1 a c'} \delta_{bd'} \\ &\quad + \mathcal{L}_{m_1 n_1 b d'} \delta_{ac'}) \bar{F}_{c'd'cd}^*(0) \Delta_{cd}^* \mu_{m_1 n_1}(\mathbf{r}) \\ &\quad + (\mathcal{L}_{m_2 n_2 a c'} \delta_{bd'} \\ &\quad + \mathcal{L}_{m_2 n_2 b d'} \delta_{ac'}) \bar{F}_{c'd'cd}^*(0) \Delta_{cd}^* \mu_{m_1 n_1}(\mathbf{r}), \end{aligned} \quad (7.9c)$$

$$\begin{aligned} \mu_{2ab}(\mathbf{r}) &= (\mathcal{L}_{m_1 n_1 a c'} \delta_{bd'} \\ &\quad + \mathcal{L}_{m_1 n_1 b d'} \delta_{ac'}) \bar{F}_{c'd'cd}(0) \Delta_{cd} \mu_{m_1 n_1}^*(\mathbf{r}) \\ &\quad + (\mathcal{L}_{m_2 n_2 a c'} \delta_{bd'} \\ &\quad + \mathcal{L}_{m_2 n_2 b d'} \delta_{ac'}) \bar{F}_{c'd'cd}(0) \Delta_{cd} \mu_{m_2 n_2}^*(\mathbf{r}), \end{aligned} \quad (7.9d)$$

where the matrices \mathcal{L} are defined in Appendix C [Eqs. (C4)]. The form of Eqs. (7.8) and (7.9) for the linear response allows us to obtain immediately an expansion through the modes of the linear problem

$$B(\omega_\alpha) \xi_\alpha = 0. \quad (7.10)$$

It is easy to show a one to one correspondence between the eigenmodes of Eqs. (7.10) and (6.4); the eigenmodes of Eq. (6.4) are given by the components $\xi_{a\alpha}$ of the eigenmodes of Eq. (7.10) [the eigenmodes of Eq. (7.10) have components ξ_{mna} as well]. The linear response $\chi(\mathbf{r}, \mathbf{r}'; \omega)$ has the form of Eq. (6.6) with $\mu_a(\mathbf{r})$ given by

$$\begin{aligned} \mu_a(\mathbf{r}) &= \xi_{1a\alpha} \mu_{1a}(\mathbf{r}) + \xi_{2a\alpha} \mu_{2a}(\mathbf{r}) + \xi_{1mn\alpha} \mu_{1mn}(\mathbf{r}) \\ &\quad + \xi_{2mn\alpha} \mu_{2mn}(\mathbf{r}), \end{aligned} \quad (7.11)$$

where the components of $\mu(\mathbf{r})$ are given by Eqs. (7.9b)–(7.9d) and the ξ_α modes are normalized according to Eq. (6.7b). Note that by setting $\mu_{m_1 n_1}(\mathbf{r}) = \mu_{m_2 n_2}(\mathbf{r}) = 0$ we recover the results of Sec. V.

VIII. DYNAMIC SELF-CONSISTENT CALCULATION OF THE GROUND STATE USING THE EFFECTIVE ACTION

The present Green-function approach makes it possible to calculate not only the dynamical behavior of quantum systems but the ground-state properties as well. These show up in Green functions with coinciding time arguments, which represent the expectation values of products of operators. The Green function perturbation theory for the ground state can be obtained as follows. We start with the ground state of the unperturbed noninteracting system at $\tau = -\infty$, and then switch the interaction adiabatically to obtain the ground state of the interacting system. By evaluating perturbatively a Green function with coinciding time arguments at a time τ_0 after the interaction is completely switched on, we obtain an expectation value of a product of operators with respect to the true ground state of the interacting system.

We now show how our formalism can be used to construct a self-consistent procedure for the ground-state reduced density matrix. In Sec. IV we have introduced the matrix $\rho^{(0)}$, which is related to $\rho^{(g)}$. The calculation starts with finding $\rho^{(0)}$ making use of Eqs. (4.5). The lhs of Eqs. (4.5) does not depend on τ and Eqs. (4.5) can be treated as a system of $M(N-M)$ equations for an $M(N-M)$ matrix $\rho^{(0)}$. The procedure for finding $\rho^{(0)}$ using the effective action formalism involves two steps: (i) evaluate S_{eff} as a function of $\rho^{(0)}$ and (ii) solve Eqs. (4.5) for $\rho^{(0)}$. Strictly speaking, this is a self-consistent procedure since the effective action calculation in the first step involves $\rho^{(0)}$, and only at the second step we find when the first step is justified.

We first carry out steps (i) and (ii) to obtain $\rho^{(0)}$ and S_{eff} . Now we are ready to evaluate $\rho^{(g)}$. Using the convention of indices [Eq. (4.2)] we can consider four types of components of $\rho^{(g)}$: $\rho_{m_1 n_2}^{(g)}$, $\rho_{m_2 n_1}^{(g)}$, $\rho_{m_1 n_1}^{(g)}$, and $\rho_{m_2 n_2}^{(g)}$. Since in the absence of the external field $\rho_{m_2 m_1}^{(g)} = x_m$, $\rho_{m_1 m_2}^{(g)} = \bar{x}_m$, and $x_m = \bar{x}_m = 0$ due to Eqs. (4.5) we have

$$\rho_{m_1 n_2}^{(g)} = \rho_{m_2 n_1}^{(g)} = 0 \quad (8.1)$$

and we need to find only the components $\rho_{m_1 n_1}^{(g)}$ and $\rho_{m_2 n_2}^{(g)}$. To do that we notice that

$$[\hat{X}_m, \hat{X}_n^\dagger] = \delta_{m_1 n_1} \hat{c}_{m_2}^\dagger \hat{c}_{n_2} - \delta_{m_2 n_2} \hat{c}_{n_1}^\dagger \hat{c}_{m_1}. \quad (8.2a)$$

Taking the expectation value of Eq. (8.2a) in the absence of the external field we obtain

$$\rho_{m_2 n_2}^{(g)} \delta_{m_1 n_1} - \rho_{n_1 m_1}^{(g)} \delta_{m_2 n_2} = \lim_{\tau \rightarrow 0^+} \langle X_{m+}(\tau) \bar{X}_n(0) \rangle_S. \quad (8.2b)$$

The rhs of Eq. (8.2b) can be easily evaluated provided S_{eff} is known. However, this calculation was already done in Sec.

V, since the Green function on the rhs of Eq. (8.2b) is related to the linear optical response of the system in the time domain. We then have

$$\rho_{m_2 n_2}^{(g)} \delta_{m_1 n_1} - \rho_{n_1 m_1}^{(g)} \delta_{m_2 n_2} = \lim_{\tau \rightarrow 0^+} \left\{ - \int \frac{d\omega}{2\pi i} e^{-i\omega\tau} \chi_{mn}^{(11)}(\omega) \right\}. \quad (8.3a)$$

Equation (8.3a), together with the relation

$$\rho_{m_1 m_1}^{(g)} + \rho_{m_2 m_2}^{(g)} = M \quad (8.3b)$$

and Eqs. (8.1), is a closed set of equations for the ground-state reduced density matrix $\rho^{(g)}$. Equation (8.3a), written in terms of the modes introduced in Sec. VI, becomes

$$\rho_{m_2 n_2}^{(g)} \delta_{m_1 n_1} - \rho_{n_1 m_1}^{(g)} \delta_{m_2 n_2} = \sum_{\alpha > 0} (\xi_{1n\alpha} \xi_{1n\alpha}^* - \xi_{2m\alpha}^* \xi_{2m\alpha}). \quad (8.4)$$

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APPENDIX A: QUADRATIC EXPANSION OF THE EFFECTIVE ACTION

In this appendix we calculate perturbatively the quadratic terms of the effective action related to the Hamiltonian of Eq. (4.1). These terms define the two-point Green function \mathcal{G} , the multiple-point Liouville-space Green functions are expressed in terms of \mathcal{G} and higher-order vertices given by $S_{\alpha_1 \dots \alpha_n}^{(n)}(\tau_1, \dots, \tau_n)$, $n \geq 3$, from Eq. (3.5). \mathcal{G} is expressed in terms of $S_{\alpha\beta}^{(2)}(\tau_1, \tau_2)$ by means of Eq. (3.4) with $S_{\alpha\beta}^{(2)}(\tau_1, \tau_2)$ substituted for $S_{0\alpha\beta}^{(2)}(\tau_1, \tau_2)$ and \mathcal{G} substituted for $G^{(0)}$.

To apply the perturbative procedure of Sec. III we partition the Hamiltonian \hat{H} [Eqs. (4.1a)] in a similar way to Eq. (3.1). We reiterate that \hat{H}_0 contains only the terms that conserve the number of electron-hole pairs (with respect to the single Slater determinant $\Omega(\rho^{(0)})$, where $\rho^{(0)}$ is determined by the ground-state reduced density matrix $\rho^{(g)}$). \hat{H}_1 , which contains the other terms, can be partitioned as

$$\hat{H}_1 = \hat{H}_1^{(1)} + \hat{H}_1^{(2)}, \quad (A1)$$

where $\hat{H}_1^{(1)}$ contains the terms that create or annihilate one electron-hole pair and $\hat{H}_1^{(2)}$ contains terms that create or annihilate two electron-hole pairs. We will evaluate the terms in S_{eff} that have the form xx or $\bar{x}\bar{x}$ and are induced by the first-order correction in $\hat{H}_1^{(2)}$ and the terms with the form $\bar{x}x$ induced by second-order corrections in $\hat{H}_1^{(1)}$ and second-order corrections in $\hat{H}_1^{(2)}$. Other corrections either vanish or appear only in higher orders. We will denote these corrections to the effective action by $S_{\text{eff}}^{(j)}$, $j = a, b, c, d$. They have the form



FIG. 3. Diagrams contributing to Δ_{mn} . (a) and (b) correspond to the first and second terms in Eq. (A7), respectively.

$$S_{\text{eff}}^{(a)}[x] = -2 \int d\tau \Delta_{ab} \bar{x}_{a+}(\tau) \bar{x}_{b-}(\tau), \quad (\text{A2a})$$

$$S_{\text{eff}}^{(b)}[x] = -2 \int d\tau \Delta_{ab}^* \bar{x}_{a+}(\tau) \bar{x}_{b-}(\tau), \quad (\text{A2b})$$

$$S_{\text{eff}}^{(c)}[x] = - \int d\tau'' d\tau' \Sigma_{ab}^{(1)}(\tau'' - \tau') \bar{x}_{a-}(\tau'') x_{b+}(\tau') - \int d\tau'' d\tau' \Sigma_{ba}^{(1)*}(\tau' - \tau'') \bar{x}_{a+}(\tau'') x_{b-}(\tau'), \quad (\text{A2c})$$

$$S_{\text{eff}}^{(d)}[x] = - \int d\tau'' d\tau' \Sigma_{ab}^{(2)}(\tau'' - \tau') \bar{x}_{a-}(\tau'') x_{b+}(\tau') - \int d\tau'' d\tau' \Sigma_{ba}^{(2)*}(\tau' - \tau'') \bar{x}_{a+}(\tau'') x_{b-}(\tau'). \quad (\text{A2d})$$

In Eqs. (A2) we always have a $+-$ combination of the Liouville-space indices of the effective action variables. The $++$ terms vanish identically (this is a general property of the Liouville-space effective action). The $--$ terms can be calculated in the same manner as the $+-$ terms presented in Eqs. (A2). Note that, strictly speaking, $S_{\text{eff}}^{(a)}$ (and $S_{\text{eff}}^{(b)} = S_{\text{eff}}^{(a)*}$) should be written in the form

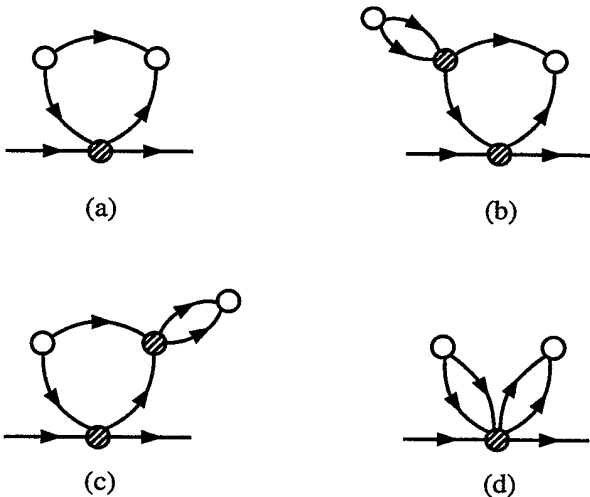


FIG. 4. Diagrams contributing to $\Sigma^{(2)}(\omega)$.

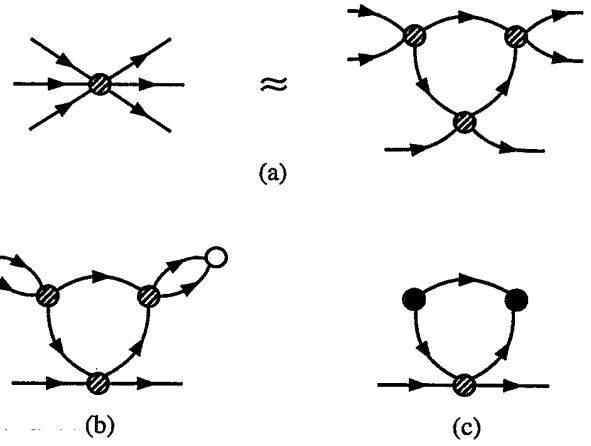


FIG. 5. Diagrams illustrating the procedure of calculating $\Sigma^{(2)}$: (a) for the approximation used for the sixth-order vertex that appears in Fig. 4(d), (b) from Fig. 4(d) under the approximation given by (a), and (c) for $\Sigma^{(2)}(\omega)$, with the dark circles denoting Δ and Δ^* .

$$S_{\text{eff}}^{(a)}[x] = -2 \int d\tau'' d\tau' \Delta_{ab}(\tau'' - \tau') \bar{x}_{a+}(\tau'') \bar{x}_{b-}(\tau'), \quad (\text{A3})$$

but we will show that within our approximation $\Delta_{ab}(\tau'' - \tau') = \Delta_{ab} \delta(\tau'' - \tau')$ and this is why we have adopted the simpler form of Eqs. (A2a) and (A2b). Using the notation of Eqs. (B6) under our approximation, Δ is proportional to V , $\Sigma^{(1)}$ to W^*W , and $\Sigma^{(2)}$ to V^*V . We will use the perturbation theory developed in Sec. III. We start by calculating Δ . Δ is given by two contributions presented diagrammatically in Fig. 3. Open circles denote type-(i) and filled circles denote type-(ii) vertices as defined in Sec. III. The second-order type-(i) vertex in Figs. 3(a) and 3(b) represents the term $S_1^{(a)}[X(\tau)]$ in the action $S_1[X(\tau)]$, which has the form (this term is induced by the Hamiltonian $\hat{H}_1^{(2)}$)

$$S_1^{(a)}[X(\tau)] = -2 \int d\tau V_{mn} \bar{X}_{m+}(\tau) \bar{X}_{n-}(\tau). \quad (\text{A4})$$

The fourth-order type-(ii) vertex in Fig. 3(b) represents the term $(S_0^{(a)})_{\text{eff}}[x(\tau)]$ in the effective action $(S_0)_{\text{eff}}[x(\tau)]$,

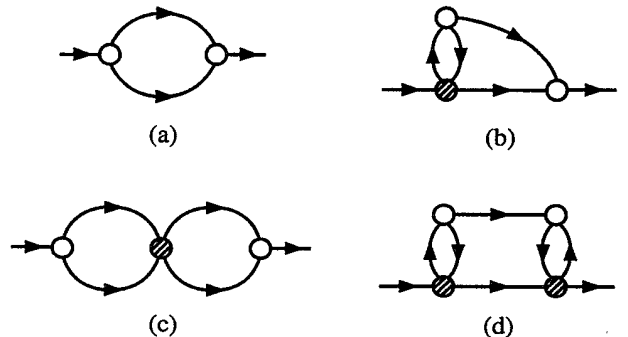


FIG. 6. Typical diagrams contributing to $\Sigma^{(1)}(\omega)$.

$$(S_0^{(a)})_{\text{eff}}[x(\tau)] = - \int d\tau d\tau' \bar{\Gamma}_{mnm'n'}^{(s)}(\tau - \tau') \times \bar{x}_{m-}(\tau) \bar{x}_{n+}(\tau) x_{m'+}(\tau') x_{n'+}(\tau'), \quad (\text{A5})$$

where the scattering matrix of two particle-hole modes $\bar{\Gamma}^{(s)}$ is evaluated in Appendix B and has the form

$$\bar{\Gamma}^{(s)}(\tau - \tau') = \int \frac{d\omega}{2\pi} \exp[-i\omega(\tau - \tau')] \bar{\Gamma}^{(s)}(\omega), \quad (\text{A6})$$

where $\bar{\Gamma}^{(s)}(\omega)$ is given by Eqs. (5.15).

The sum of contributions of both diagrams in Fig. 3 gives

$$\Delta_{mn} = V_{mn} + 2\bar{\Gamma}_{mnm'n'}^{(s)}(0) \bar{F}_{m'n'an}(0) V_{ab}. \quad (\text{A7})$$

The first and second terms on the rhs of Eq. (A7) are represented diagrammatically in Figs. 3(a) and 3(b), respectively. Substituting $\bar{\Gamma}^{(s)}$ from Eq. (5.15a) into Eq. (A7), we obtain Δ in a form given by Eqs. (5.3) and (5.9). We will now evaluate $\Sigma^{(2)}$, which is given as a sum of contributions presented diagrammatically in Fig. 4. Figure 4(d) contains a type-(ii) six-order vertex that involves the three-mode (particle-hole) states of the Hamiltonian. To avoid dealing with three-mode states, we will make an approximation. The

six-order vertex can be expressed in terms of the boson scattering matrix $\bar{\Gamma}^{(b)}$ in the form of an infinite number of contributions related to the number of the two-particle scattering events in the propagation of three particles (starting with three scattering processes). We invoke an approximation that takes into account only three scattering events. The six-order vertex can then be expressed in terms of the four-order vertex as shown in Fig. 5(a). The diagram in Fig. 4(d) then adopts the form presented in Fig. 5(b) and the sum of four diagrams in Fig. 4 is given by the diagram in Fig. 5(c), where a dark circle denotes Δ_{mn} . Note that the expression related to the diagram in Fig. 5(c) can be obtained from that of the diagram in Fig. 4(a) if we substitute Δ_{mn} instead of V_{mn} ; this expression gives $\Sigma^{(2)}$ in the form of Eq. (5.10c).

The expression for $\Sigma^{(1)}$ can be presented diagrammatically. Typical diagrams are shown in Fig. 6. The third-order type-(i) vertices are obtained by recasting $H_1^{(1)}$ through the third-order terms in the operators \hat{X}_m . However, such a calculation, though feasible, is rather inconvenient since it yields a simple expression in the form of a sum of a large number of terms. To avoid it we use the following trick. One can easily find $\Sigma^{(1)}$ from the correction to the one-particle Green function proportional to W^*W . The latter can be calculated using the bosonization scheme presented in Appendix D. This leads to the equation

$$-i \int d\tau d\tau' \langle X_{m+}(t) \bar{X}_{m'-}(\tau) \rangle_{S_0} \langle X_{n'+}(\tau') \bar{X}_n(t') \rangle_{S_0} \Sigma_{m'n'}^{(1)}(\tau - \tau') = - \int d\tau d\tau' \langle \hat{b}_{m+}(t) \hat{b}_{m'-}^\dagger(\tau) \hat{b}_{a+}(\tau) \hat{b}_{b+}(\tau) \hat{b}_{a'+}^\dagger(\tau') \hat{b}_{b'-}^\dagger(\tau') \hat{b}_{n'+}(\tau') \hat{b}_{n-}^\dagger(\tau') \rangle_{H_0} W_{abm}^* W_{a'b'n}. \quad (\text{A8a})$$

Making use of

$$\langle \hat{b}_{m+}(t) \hat{b}_{m'-}^\dagger(\tau) \hat{b}_{a+}(\tau) \hat{b}_{b+}(\tau) \hat{b}_{a'+}^\dagger(\tau') \hat{b}_{b'-}^\dagger(\tau') \hat{b}_{n'+}(\tau') \hat{b}_{n-}^\dagger(\tau') \rangle_{H_0} = \langle X_{m+}(t) \bar{X}_{m'-}(\tau) \rangle_{S_0} \langle X_{n'+}(\tau') \bar{X}_n(t') \rangle_{S_0} \langle \hat{b}_{a+}(\tau) \hat{b}_{b+}(\tau) \hat{b}_{a'+}^\dagger(\tau') \hat{b}_{b'-}^\dagger(\tau') \rangle_{H_0}, \quad (\text{A8b})$$

we obtain From Eq. (A8a)

$$\Sigma_{ab}^{(1)}(\tau - \tau') = -i W_{mna}^* W_{m'n'b} \langle \hat{b}_{m+}(\tau) \hat{b}_{n+}(\tau) \hat{b}_{m'+}^\dagger(\tau') \hat{b}_{n'-}^\dagger(\tau') \rangle_{H_0}. \quad (\text{A9})$$

Substituting finally Eq. (B7) into Eq. (A9), we obtain $\Sigma^{(1)}$ in the form of Eq. (5.10b).

APPENDIX B: BOSON REPRESENTATION OF THE HAMILTONIAN AND GREEN FUNCTIONS

In this appendix we express the Hamiltonian of Eq. (4.1) and the operators \hat{X}_m defined by Eqs. (4.3b) in terms of boson operators, and calculate the two-mode scattering matrix $\bar{\Gamma}^{(s)}$. The bosonization is a very effective tool for perturbative calculations of S_{eff} since it makes it possible to apply conventional Green-function techniques starting with free-particle Hamiltonians. We emphasize that bosonization is not essential to the effective action approach.

Bosonization schemes have been initially introduced for spin operators [46–48] and then generalized for bilinear combinations of fermion operators [12]. Bosonization proce-

dures are not unique: for a given operator algebra, different realizations in terms of boson operators are possible [49]. Two approaches for bosonization are known [12]. The first is based on obtaining expressions for given operators in terms of boson operators that satisfy desired commutation relations. The other approach, which will be used here, involves an immersion of the representation space of a boson (oscillator) algebra.

Let \mathcal{V} be the space of states of a quantum system and \mathcal{A} be the algebra of operators acting on \mathcal{V} . We assume that the algebra \mathcal{A} is generated by operators \hat{X}_m . Let \mathcal{U} be the space of states of a set of oscillators and \mathcal{B} its operator algebra, generated by boson operators $\hat{b}_n, \hat{b}_n^\dagger$, which obey

the commutation relations $[\hat{b}_n, \hat{b}_m^\dagger] = \delta_{mn}$. Bosonization is a decomposition $\mathcal{U} = \mathcal{U}_0 \oplus \mathcal{U}_1$, where \mathcal{U}_1 is orthogonal to \mathcal{U}_0 with a linear isomorphism $f: \mathcal{V} \rightarrow \mathcal{U}_0$ and an algebra morphism $\hat{f}: \mathcal{A} \rightarrow \mathcal{B}$ with the following properties: for arbitrary $\hat{X} \in \mathcal{A}$

$$\hat{f}(\hat{X})\mathcal{U}_0 \subset \mathcal{U}_0, \quad \hat{f}(\hat{X})\mathcal{U}_1 \subset \mathcal{U}_1 \quad (\text{B1})$$

and for arbitrary $v \in \mathcal{V}$

$$f(\hat{X}(v)) = \hat{f}(\hat{X})f(v). \quad (\text{B2})$$

In other words, $f(\hat{X})$ is a boson representation of an operator \hat{X} and f immerses \mathcal{V} into \mathcal{U} . Equations (B1) and (B2) imply that \hat{X} acts on \mathcal{V} in the same way that the boson representation $\hat{f}(\hat{X})$ of \hat{X} acts on \mathcal{U}_0 , which is a replica of \mathcal{V} immersed into \mathcal{U} . When it is not confusing we will write \hat{X} instead of $\hat{f}(\hat{X})$. We now define the bosonization of our system. Let $\hat{b}_m, \hat{b}_m^\dagger$ be a set of boson operators, where the index m is defined by Eq. (4.2). The ground state of the oscillator system is denoted $|\Omega\rangle$: $\hat{b}_m|\Omega\rangle = 0$. We then define $f(\Omega(\rho^{(0)})) \equiv |\Omega\rangle$ and for each set $m_1^{(1)}, \dots, m_1^{(j)}, m_2^{(1)}, \dots, m_2^{(j)}$,

$$f[\hat{c}_{m_1}^{\dagger(1)} \hat{c}_{m_2}^{\dagger(1)} \dots \hat{c}_{m_1}^{\dagger(j)} \hat{c}_{m_2}^{\dagger(j)}] |\Omega(\rho^{(0)})\rangle \\ \equiv \frac{1}{\sqrt{j!}} \sum_p (-1)^{s(p)} \hat{b}_{p^{(1)}(m_1)m_2^{(1)}}^\dagger \dots \hat{b}_{p^{(j)}(m_1)m_2^{(j)}}^\dagger |\Omega\rangle, \quad (\text{B3})$$

where Σ_p means a sum over $j!$ permutations of $m_1^{(1)}, \dots, m_1^{(j)}$ and $(-1)^{s(p)}$ is the sign of a permutation p . Equation (B3) defines \mathcal{U}_0 , \mathcal{U}_1 , and f . The boson representation for operators may be found by applying the conditions of Eqs. (B1) and (B2).

The operators \hat{X}_m^\dagger and \hat{X}_m can be presented in the form of expansions in $\hat{b}_n^\dagger, \hat{b}_n$ written in the normal ordered form (annihilation operators on the right, creation operators on the left). We have

$$\hat{X}_m^\dagger = \hat{b}_{m_1 m_2}^\dagger + \left(\frac{1}{\sqrt{2}} - 1\right) \hat{b}_{m_1 m_2}^\dagger \hat{b}_{n_1 n_2}^\dagger \hat{b}_{n_1 n_2} \\ - \frac{1}{\sqrt{2}} \hat{b}_{m_1 m_2}^\dagger \hat{b}_{n_1 n_2}^\dagger \hat{b}_{n_1 n_2} + \dots, \quad (\text{B4a})$$

$$\hat{X}_m = \hat{b}_{m_1 m_2} + \left(\frac{1}{\sqrt{2}} - 1\right) \hat{b}_{n_1 n_2}^\dagger \hat{b}_{n_1 n_2} \hat{b}_{m_1 m_2} \\ - \frac{1}{\sqrt{2}} \hat{b}_{n_1 n_2}^\dagger \hat{b}_{n_1 n_2} \hat{b}_{m_1 m_2} + \dots. \quad (\text{B4b})$$

We also have for the Hamiltonian \hat{H}_0 , $\hat{H}_0 = \hat{H}_0^{(0)} + \hat{H}_0^{(\text{int})}$ with

$$\hat{H}_0^{(0)} = L_{mn} \hat{b}_m^\dagger \hat{b}_n, \quad \hat{H}_0^{(\text{int})} = U_{mnm'n'} \hat{b}_m^\dagger \hat{b}_n^\dagger \hat{b}_{m'} \hat{b}_{n'} \quad (\text{B5})$$

and for \hat{H}_1 , $\hat{H}_1 = \hat{H}_1^{(1)} + \hat{H}_1^{(2)}$ with

$$\hat{H}_1^{(1)} = W_{mnp} \hat{b}_m^\dagger \hat{b}_n^\dagger \hat{b}_p + W_{mnp}^* \hat{b}_p \hat{b}_n \hat{b}_m + \dots, \quad (\text{B6a})$$

$$\hat{H}_1^{(2)} = V_{mn} \hat{X}_m^\dagger \hat{X}_n^\dagger + V_{mn}^* \hat{X}_m \hat{X}_n, \quad (\text{B6b})$$

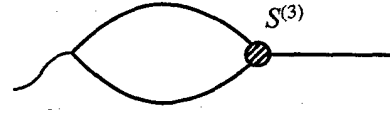


FIG. 7. Diagram representation for the Green functions of Eqs. (C1a) and (C1b) in terms of the effective action. The hatched circle denotes $S^{(3)}$ defined by Eq. (3.5).

where the matrices H , U , W , and V are defined in Sec. V by Eqs. (5.8) and (5.16)–(5.18).

We can now introduce the boson-boson scattering matrix $\bar{\Gamma}_{mnm'n'}^{(b)}(\Omega)$ with respect to the Hamiltonian \hat{H}_0

$$\langle \hat{b}_{m_+}(t) \hat{b}_{n_+}(t) \hat{b}_{m'_-}^\dagger(t') \hat{b}_{n'_-}^\dagger(t') \rangle_{\hat{H}_0} \\ = i \int \frac{d\Omega}{2\pi} \exp\{-i\Omega(t-t')\} \{2\bar{F}_{mnm'n'}(\Omega) \\ + 4\bar{F}_{mnab}(\Omega) \bar{\Gamma}_{aba'b'}^{(b)}(\Omega) \bar{F}_{a'b'm'n'}(\Omega)\}, \quad (\text{B7})$$

where $\bar{F}(\Omega)$ is given by Eqs. (5.13). $\bar{\Gamma}^{(b)}$ can be evaluated using the representation of Eq. (B5) and \hat{H}_0 and standard perturbation theory for Green functions [42,44]. The exact value of $\bar{\Gamma}^{(b)}$ is given by the sum of ladder contributions (see [5,30]) and $\bar{\Gamma}^{(b)}$ has the form of Eq. (5.14).

The two-mode scattering matrix $\bar{\Gamma}^{(s)}$ is different from $\bar{\Gamma}^{(b)}$ since it takes into account effects of nonboson statistics in the mode scattering. To obtain $\bar{\Gamma}^{(s)}$ we make use of the fact that $\bar{\Gamma}^{(s)}$ is a part of the effective action $(S_0)_{\text{eff}}$ according to Eq. (A5) and hence appears in the four-point correlation functions of \hat{X} and \hat{X}^\dagger evolving with the Hamiltonian \hat{H}_0 . On the other hand, these correlation functions can be expressed through $\bar{\Gamma}^{(b)}$ using the bosonization scheme. The simplest way to obtain $\bar{\Gamma}^{(s)}$ through $\bar{\Gamma}^{(b)}$ is to compare two expressions for the Green function $\langle X_{m_+}(\tau) X_{n_+}(\tau) \bar{X}_{m'_-}(\tau') \bar{X}_{n'_-}(\tau') \rangle_{S_0}$. This yields

$$\langle X_{m_+}(\tau) X_{n_+}(\tau) \bar{X}_{m'_-}(\tau') \bar{X}_{n'_-}(\tau') \rangle_{S_0} \\ = \frac{1}{2} (I-P)_{mnan} (I-P)_{a'b'm'n'} \\ \times \langle \hat{b}_{a_+}(\tau) \hat{b}_{b_+}(\tau) \hat{b}_{a'_-}^\dagger(\tau') \hat{b}_{b'_-}^\dagger(\tau') \rangle_{H_0}, \quad (\text{B8})$$

where P is defined by Eq. (5.11). Expressing the lhs of Eq. (B8) in terms of $\bar{\Gamma}^{(s)}$ and the rhs of Eq. (B8) in terms of $\bar{\Gamma}^{(b)}$ [Eq. (B7)], we obtain the operator relation

$$\frac{1}{2} (I-P) \{2\bar{F}(\omega) + 4\bar{F}(\omega) \bar{\Gamma}^{(b)}(\omega) \bar{F}(\omega)\} (I-P) \\ = 2\bar{F}(\omega) + 4\bar{F}(\omega) \bar{\Gamma}^{(s)}(\omega) \bar{F}(\omega). \quad (\text{B9})$$

Since the expression in the curly brackets in Eq. (B9) is the

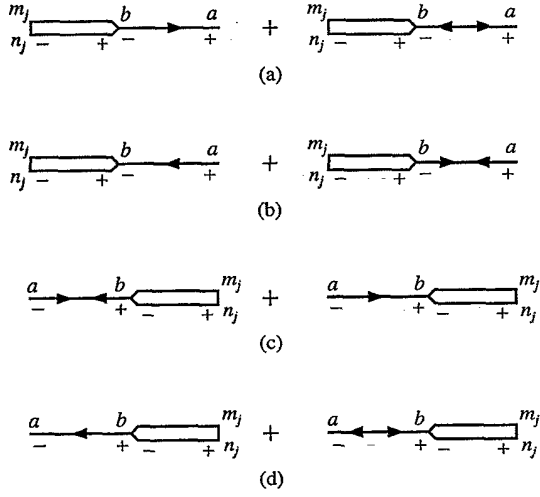


FIG. 8. Diagram representation of Eqs. (C3). The single lines stand for \mathcal{G} , the double lines for \mathcal{F} .

two-particle Green function of the Hamiltonian \hat{H}_0 , it commutes with $I-P$ (note that the two terms do not commute separately with $I-P$). Making use of this commutation and the relation $P^2=I$, we obtain Eqs. (5.15).

APPENDIX C: CALCULATION OF THE GREEN FUNCTIONS

In this appendix we evaluate the Green functions $\langle \hat{X}_{a,b,\alpha}(\tau') \hat{X}_{m,\beta}(\tau) \rangle$ and $\langle \hat{X}_{a,b,\alpha}(\tau') \hat{X}_{a',b',\beta}(\tau) \rangle$, where $j, k=1,2$ and $\hat{X}_{a,b_j} \equiv \hat{c}_{a_j}^\dagger \hat{c}_{b_j}$, and the operators $\hat{X}_m, \hat{X}_m^\dagger$ are defined by Eq. (4.3b). The operators \hat{X}_{a,b_j} are the particle-particle and hole-hole components of the reduced density matrix. We need these Green functions to express the components of the linear response that are related to the particle-particle and hole-hole as well as particle-hole components of the reduced density matrix in the expansion of the polarization operator. For the linear response we need these Green functions for $\alpha=+, \beta=-$ or $\alpha=-, \beta=+$. We introduce the Green functions (see Fig. 7)

$$\langle \hat{X}_{a\alpha}(\tau') \hat{X}_{m,n_j,\beta}(\tau) \rangle = i \int \frac{d\omega}{2\pi} e^{-i\omega(\tau'-\tau)} \mathcal{G}_{am,n_j,\alpha\beta}^{(1)}(\omega), \quad (\text{C1a})$$

$$\langle \hat{X}_{a\alpha}^\dagger(\tau') \hat{X}_{m,n_j,\beta}(\tau) \rangle = i \int \frac{d\omega}{2\pi} e^{-i\omega(\tau'-\tau)} \mathcal{G}_{am,n_j,\alpha\beta}^{(2)}(\omega), \quad (\text{C1b})$$

$$\langle \hat{X}_{a,b_j,\alpha}(\tau') \hat{X}_{m_k,n_k,\beta}(\tau) \rangle = i \int \frac{d\omega}{2\pi} e^{-i\omega(\tau'-\tau)} \mathcal{G}_{a,b_j,m_k,n_k,\alpha\beta}(\omega), \quad (\text{C1c})$$

with $j, k=1,2$.

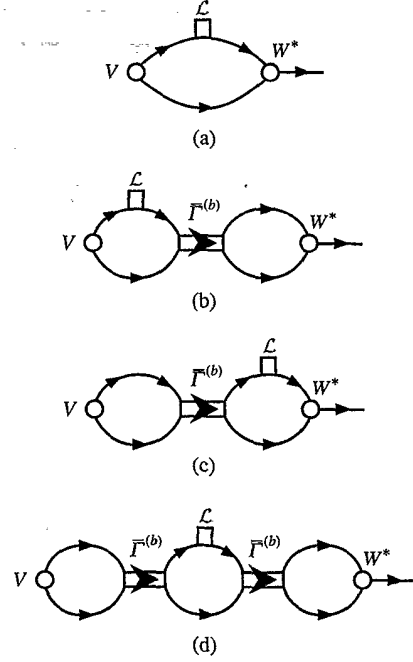


FIG. 9. (a)–(d) Four contributions to the Green function on the rhs of Eq. (C7). The open circles denote V and W^* .

$$\langle \hat{X}_{m\alpha}(\tau') \hat{X}_{n\beta}^\dagger(\tau) \rangle = i \int \frac{d\omega}{2\pi} e^{-i\omega(\tau'-\tau)} \mathcal{G}_{mn\alpha\beta}^{(11)}(\omega), \quad (\text{C2a})$$

$$\langle \hat{X}_{m\alpha}^\dagger(\tau') \hat{X}_{n\beta}(\tau) \rangle = i \int \frac{d\omega}{2\pi} e^{-i\omega(\tau'-\tau)} \mathcal{G}_{mn\alpha\beta}^{(22)}(\omega), \quad (\text{C2b})$$

$$\langle \hat{X}_{m\alpha}(\tau') \hat{X}_{n\beta}(\tau) \rangle = i \int \frac{d\omega}{2\pi} e^{-i\omega(\tau'-\tau)} \mathcal{G}_{mn\alpha\beta}^{(12)}(\omega), \quad (\text{C2c})$$

$$\langle \hat{X}_{m\alpha}^\dagger(\tau') \hat{X}_{n\beta}^\dagger(\tau) \rangle = i \int \frac{d\omega}{2\pi} e^{-i\omega(\tau'-\tau)} \mathcal{G}_{mn\alpha\beta}^{(21)}(\omega). \quad (\text{C2d})$$

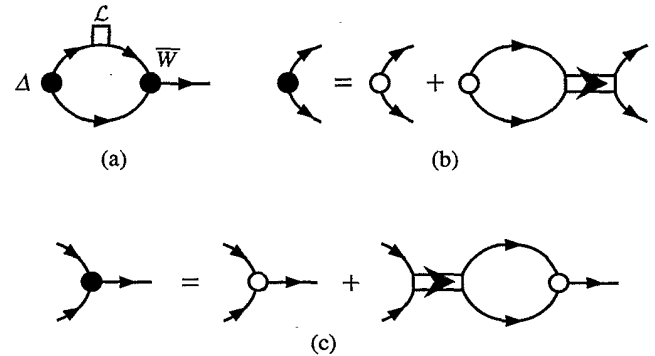


FIG. 10. (a) Sum of four contributions of Fig. 9 presented in a form of a single diagram. The dark circles stand for Δ and \bar{W} . (b) Diagram representation for Δ . (c) Diagram representation for \bar{W} .

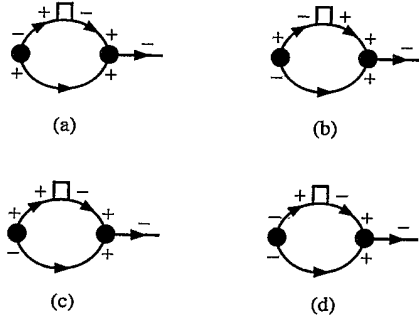


FIG. 11. (a)–(c) Three possible combinations of the time-loop related indices $\alpha = \pm$ on the diagram shown in Fig. 10(a). (d) The sum of diagrams (a)–(c) combined to a single diagram.

It follows from the correlation function expressions for the linear response $\mathcal{F}_{mn+}^{(ij)}(\omega) = \chi_{mn}^{(ij)}(\omega)$, where $\chi_{mn}^{(ij)}(\omega)$ is defined in Sec. V [Eqs. (5.7)].

To evaluate the Green functions of Eqs. (C1a) and (C1b) for $\alpha = +$, $\beta = -$ or $\alpha = -$, $\beta = +$ we make use of the commutation relations for the \hat{X} operators, which yield $[\hat{X}_n, \hat{X}_m^\dagger] = \delta_{n_1 m_1} \hat{X}_{n_2 m_2} - \delta_{m_2 n_2} \hat{X}_{m_1 n_1}$, and express them in terms of the effective action. However, it is more convenient to use the following trick. The Green functions can be presented as

$$\begin{aligned} \mathcal{F}_{am,n_j+}^{(1)}(\omega) &= \mathcal{F}_{ab+}^{(11)}(\omega) \mathcal{F}_{bm,n_j+}^{(2)}(\omega) \\ &+ \mathcal{F}_{ab+}^{(12)}(\omega) \mathcal{F}_{bm,n_j+}^{(1)}(\omega), \end{aligned} \quad (\text{C3a})$$

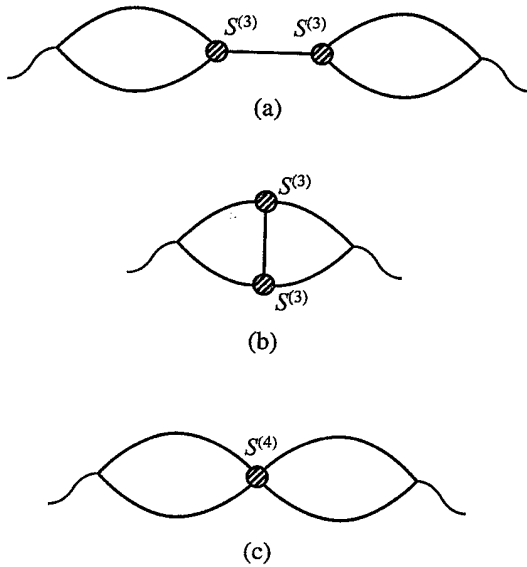


FIG. 12. (a)–(c) Diagram representation of three contributions to the Green function defined by Eq. (C1c) expressed in terms of the effective action.

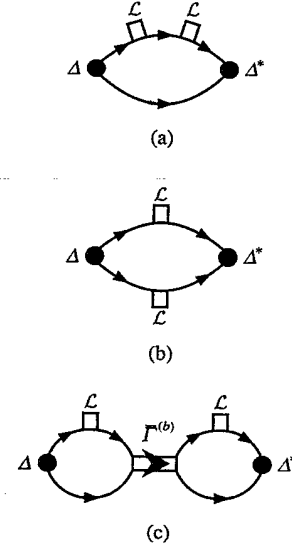


FIG. 13. Three contributions of $\mathcal{F}^{(3)}$ in Eq. (C13a) given by Eqs. (C13b)–(C13d).

$$\begin{aligned} \mathcal{F}_{am,n_j+}^{(2)}(\omega) &= \mathcal{F}_{ab+}^{(21)}(\omega) \mathcal{F}_{bm,n_j+}^{(2)}(\omega) \\ &+ \mathcal{F}_{ab+}^{(22)}(\omega) \mathcal{F}_{bm,n_j+}^{(1)}(\omega), \end{aligned} \quad (\text{C3b})$$

$$\begin{aligned} \mathcal{F}_{am,n_j-}^{(2)}(\omega) &= \mathcal{F}_{m_j n_j b+}^{(1)}(-\omega) \mathcal{F}_{ba+}^{(21)}(-\omega) \\ &+ \mathcal{F}_{m_j n_j b+}^{(2)}(-\omega) \mathcal{F}_{ba+}^{(11)}(-\omega), \end{aligned} \quad (\text{C3c})$$

$$\begin{aligned} \mathcal{F}_{am,n_j-}^{(1)}(\omega) &= \mathcal{F}_{m_j n_j b+}^{(1)}(-\omega) \mathcal{F}_{ba+}^{(22)}(-\omega) \\ &+ \mathcal{F}_{m_j n_j b+}^{(2)}(-\omega) \mathcal{F}_{ba+}^{(12)}(-\omega) \end{aligned} \quad (\text{C3d})$$

(see Fig. 8 for the diagrammatic representation).

Equations (C3) express the desired Green functions in terms of $\chi_{mn}^{(ij)}(\omega)$, which are calculated in Sec. V, and the matrices \mathcal{F} . The matrices \mathcal{F} can be evaluated by expanding Eqs. (C3) in W and V and the expansions can be carried out by applying the bosonization technique. We will show how to calculate $\mathcal{F}_{bm,n_j+}^{(2)}(\omega)$ using Eq. (C3a); other components can be obtained in the same manner by making use of Eqs. (C3b)–(C3d).

The expansion of $\mathcal{F}^{(1)}$ and $\mathcal{F}^{(2)}$ starts with W . Consider Eqs. (C3a) and (C3b) in this order. We will show later that the W term in $\mathcal{F}^{(1)}$ and $\mathcal{F}^{(2)}$ is zero. Since the corrections to $\mathcal{F}^{(ij)}$ in W start with W^*W , we should take $\mathcal{F}^{(ij)}$ in the zeroth order and this leads to $\mathcal{F} = 0$ for terms proportional to W . This means that we should take into account the terms in $\mathcal{F} \propto WV^*$. To do this we evaluate $\mathcal{F}^{(1)}$ in this order and then we take $\mathcal{F}^{(11)}$ and $\mathcal{F}^{(12)}$ in zeroth order, i.e., $\mathcal{F}_{+-}^{(11)} = G$ [G is given by Eq. (5.12)], $\mathcal{F}_{+-}^{(12)} = 0$. Equation (C3a) then becomes $\mathcal{F}_{am,n_j+}^{(1)}(\omega) = G_{ab}(\omega) \mathcal{F}_{bm,n_j+}^{(2)}(\omega)$, where \mathcal{F} denotes the WV^* term in the expansion of $\mathcal{F}^{(1)}$. The expansion can be obtained by applying the boson representation to Eq. (3.2)

and expanding the exponent on its rhs. The correlation functions of the boson operators obtained after the expansion of the exponent are defined with respect to the Hamiltonian \hat{H}_0 . The operators \hat{X}_{m,n_j} ($j=1,2$) assume the following form in terms of the boson operators $\hat{b}_m, \hat{b}_m^\dagger$: $\hat{X}_{m,n_j} = \mathcal{L}_{m,n_j,pq} \hat{b}_p^\dagger \hat{b}_q$ with

$$\begin{aligned} \mathcal{L}_{m_1 n_1 p q} &\equiv \delta_{m_1 p_1} \delta_{n_1 q_1} \delta_{p_2 q_2}, \\ \mathcal{L}_{m_2 n_2 p q} &\equiv \delta_{m_2 p_2} \delta_{n_2 q_2} \delta_{m_1 n_1}. \end{aligned} \quad (C4)$$

The contribution to $\mathcal{F}_{+-}^{(1)}$ proportional to W is denoted by $\tilde{\mathcal{F}}_{+-}^{(1)}$ and has the form

$$\begin{aligned} \tilde{\mathcal{F}}_{am,n_j+-}^{(1)}(\omega) &= -i \mathcal{L}_{m,n_j,pq} W_{svu} \int d\tau d\tau' \exp(i\omega\tau) \\ &\times \langle [\hat{b}_p^\dagger(0) \hat{b}_q(0)]_{-\hat{b}_{a+}(\tau)} \\ &\times [\hat{b}_s^\dagger(\tau') \hat{b}_v^\dagger(\tau') \hat{b}_u(\tau')]_{-\hat{H}_0} \rangle. \end{aligned} \quad (C5)$$

The correlation function on the rhs of Eq. (C5) can be calculated using the diagram technique for boson systems with interactions. It is easy to show that each diagram for the correlation function of Eq. (C5) contains a loop and hence it vanishes since the ground state of \hat{H}_0 contains no particles and we get

$$\tilde{\mathcal{F}}_{am,n_j+-}^{(1)}(\omega) \equiv 0. \quad (C6)$$

The expression for $\tilde{\mathcal{F}}_{+-}^{(1)}(\omega)$ is

$$\begin{aligned} \tilde{\mathcal{F}}_{am,n_j+-}^{(1)}(\omega) &= -\mathcal{L}_{m,n_j,pq} \frac{1}{\sqrt{2}} (I-P)_{cdc'd'} V_{c'd'} W_{svu}^* \\ &\times \int d\tau d\tau' d\tau'' \exp(i\omega\tau) \langle [\hat{b}_p^\dagger(0) \hat{b}_q(0)]_{-\hat{b}_{a+}(\tau)} \\ &\times [\hat{b}_c^\dagger(\tau') \hat{b}_d^\dagger(\tau')]_{-\hat{H}_0} - [\hat{b}_u^\dagger(\tau'') \hat{b}_s(\tau'') \hat{b}_v(\tau'')]_{-\hat{H}_0} \rangle. \end{aligned} \quad (C7)$$

The correlation function on the rhs of Eq. (C7) can be calculated using the diagrammatic technique for boson systems by taking into account all the diagrams that do not contain loops. The summation result is presented in Figs. 9(a)–9(d) in diagrammatic form as a sum of four contributions (single and double lines stand for G and $\bar{\Gamma}^{(b)}$ and open circles denote V and W^* . The sum of these four contributions can be presented in the form of one diagram [see Fig. 10(a)]. The filled circles stand for Δ_{mn} and $\bar{W}_{a,mn}(\omega)$ [they are presented diagrammatically in Figs. 10(b) and 10(c)]; Δ_{mn} is given by Eqs. (5.3) and (5.9) and $\bar{W}_{a,mn}(\omega)$ has the form

$$\bar{W}_{a,mn}(\omega) = W_{mna}^* + 2W_{pqaf}^* \bar{F}_{pqm'n'}(\omega) \bar{\Gamma}_{m'n'mn}^{(b)}(\omega). \quad (C8)$$

All possible values of the Liouville-space related indices are shown in Figs. 11(a)–11(c). Making use of $G_{++}(\omega)$

$= \frac{1}{2}[G(\omega) - G^*(\omega)]$, we end up with one diagram, shown in Fig. 11(d), and we finally obtain the expression for $\mathcal{F}_{+-}^{(2)}$ combining Eq. (C7) and Fig. 11(d):

$$\begin{aligned} \mathcal{F}_{am,n_j+-}^{(2)}(\omega) &= \mathcal{L}_{m,n_j,pq} \Delta_{cd} \sqrt{2} \bar{W}_{a,uv}(\omega) \\ &\times \int \frac{d\omega'}{2\pi i} G_{vd}(\omega') G_{qc}(-\omega') G_{up}(\omega - \omega'). \end{aligned} \quad (C9)$$

The other components of \mathcal{F} can be obtained in the same way.

The Green function \mathcal{F} from Eq. (C1c) can be expressed in terms of the Green functions $\mathcal{F}^{(ij)}$ of Eqs. (C2) and the functions $S^{(3)}$ and $S^{(4)}$ introduced in Eq. (3.5). \mathcal{F} is given in the form of a sum of three contributions presented diagrammatically in Figs. 12(a)–12(c). The expansion of the contributions presented in Figs. 12(a) and 12(b) in V and W begins with the term proportional to $V^* V W^* W$ and the expansion of the contribution from Fig. 12(c) starts with $V^* V$ and does not have $W^* W$ terms. This means that the first two contributions have a higher order than the third; however, the first contribution [Fig. 12(a)] contains resonant terms related to the Green function $\mathcal{F}^{(ij)}$ connecting two vertices. For this reason we will neglect the second contribution [Fig. 12(b)] and evaluate the third contribution in the first nonvanishing order, i.e., $V^* V$ terms. Summarizing, under our approximation \mathcal{F} is given as

$$\mathcal{F}_{a_j b_j m_k n_k \alpha \beta}(\omega) = \mathcal{F}_{a_j b_j m_k n_k \alpha \beta}^{(a)}(\omega) + \mathcal{F}_{a_j b_j m_k n_k \alpha \beta}^{(c)}(\omega). \quad (C10)$$

To evaluate $\mathcal{F}^{(a)}$ we will express it in terms of the matrices \mathcal{F} introduced in Appendix A. $\mathcal{F}^{(c)}$ will be obtained by evaluating the lhs of Eq. (C1c) directly by using the bosonization technique and taking into account the $V^* V$ term only.

To obtain an expression for $\mathcal{F}^{(a)}$ we make use of the definition of \mathcal{F} [Eqs. (C3) and Fig. 8] and the representation of $\mathcal{F}^{(a)}$ given by Fig. 12(a), which yields

$$\begin{aligned} \mathcal{F}_{m_i n_i m'_j n'_j +-}^{(a)}(\omega) &= \mathcal{F}_{m_i n_i a' +-}^{(2)}(\omega) \mathcal{F}_{a' a +-}^{(11)}(\omega) \mathcal{F}_{a m'_j n'_j +-}^{(2)}(\omega) \\ &+ \mathcal{F}_{m_i n_i a' +-}^{(1)}(\omega) \mathcal{F}_{a' a +-}^{(22)}(\omega) \mathcal{F}_{a m'_j n'_j +-}^{(1)}(\omega) \\ &+ \mathcal{F}_{m_i n_i a' +-}^{(2)}(\omega) \mathcal{F}_{a' a +-}^{(12)}(\omega) \mathcal{F}_{a m'_j n'_j +-}^{(1)}(\omega) \\ &+ \mathcal{F}_{m_i n_i a' +-}^{(1)}(\omega) \mathcal{F}_{a' a +-}^{(21)}(\omega) \mathcal{F}_{a m'_j n'_j +-}^{(2)}(\omega). \end{aligned} \quad (C11)$$

Taking \mathcal{F} in the form of Eq. (C9) and $\mathcal{F}^{(ij)} = \chi^{(ij)}$ with $\chi^{(ij)}$ given by Eqs. (5.7), we obtain $\mathcal{F}^{(a)}$ from Eq. (C11).

To obtain $\mathcal{F}^{(c)}$ we apply the boson representation to Eq. (3.2), expand the exponent up to the $V^* V$ terms, and make use of the representation of Eq. (C4) for operators \hat{X}_{m,n_j} . This yields

$$\begin{aligned} \mathcal{F}_{m_i n_i m'_j n'_j}^{(c)} + (\omega) &= \frac{i}{2} \mathcal{L}_{m_i n_i a b} \mathcal{L}_{m'_j n'_j a' b'} (I-P)_{c' d' c d} (I-P)_{p' q' p q} V_{c' d'} V_{p' q'}^* \\ &\times \int d\tau d\tau' d\tau'' \exp(i\omega\tau) \langle [\hat{b}_a^\dagger(\tau) \hat{b}_b(\tau)]_+ [\hat{b}_{a'}(0) \hat{b}_{b'}(0)]_- [\hat{b}_c^\dagger(\tau') b_d^\dagger(\tau')]_- [\hat{b}_p(\tau'') \hat{b}_q(\tau'')]_- \rangle_{\mathbb{H}_0}. \end{aligned} \quad (C12)$$

We evaluate the correlation function on the rhs of Eq. (C12) using the technique for systems of interacting bosons, taking into account all perturbative terms related to diagrams without loops, to obtain the exact value. The result has the form of three contributions presented diagrammatically in Figs. 13(a)–13(c) using the notation of Fig. 10. $\mathcal{F}^{(c)}$ adopts the form

$$\mathcal{F}_{m_i n_i m'_j n'_j}^{(c)} + (\omega) = \mathcal{F}_{m_i n_i m'_j n'_j}^I(\omega) + \mathcal{F}_{m_i n_i m'_j n'_j}^{II}(\omega) + \mathcal{F}_{m_i n_i m'_j n'_j}^{III}(\omega) \quad (C13a)$$

with

$$\begin{aligned} \mathcal{F}_{m_i n_i m'_j n'_j}^I(\omega) &\equiv -2 \int \frac{d\omega'}{2\pi i} G_{qd}(\omega') G_{b'c}(-\omega') G_{ba'}(-\omega' + \omega) G_{pa}(-\omega') \mathcal{L}_{m'_j n'_j a' b'} \mathcal{L}_{m_i n_i ab} \Delta_{cd} \Delta_{pq}^* \\ &+ 2 \int \frac{d\omega'}{2\pi i} G_{qd}^*(\omega') G_{ac}^*(-\omega') G_{a'b}^*(-\omega' - \omega) G_{pb'}^*(-\omega') \mathcal{L}_{m'_j n'_j b' a'} \mathcal{L}_{m_i n_i ba} \Delta_{cd} \Delta_{pq}^*, \end{aligned} \quad (C13b)$$

$$\begin{aligned} \mathcal{F}_{m_i n_i m'_j n'_j}^{II}(\omega) &\equiv -2 \int \frac{d\omega'}{2\pi i} G_{bd}(\omega') G_{qa}^*(\omega' - \omega) G_{b'c}(-\omega') G_{pa'}(-\omega' + \omega) \mathcal{L}_{m'_j n'_j a' b'} \mathcal{L}_{m_i n_i ab} \Delta_{cd} \Delta_{pq}^* \\ &+ 2 \int \frac{d\omega'}{2\pi i} G_{qa}^*(\omega') G_{pa'}^*(-\omega') G_{bd}(\omega' + \omega) G_{b'c}^*(-\omega' - \omega) \mathcal{L}_{m'_j n'_j a' b'} \mathcal{L}_{m_i n_i ab} \Delta_{cd} \Delta_{pq}^*, \end{aligned} \quad (C13c)$$

$$\begin{aligned} \mathcal{F}_{m_i n_i m'_j n'_j}^{III}(\omega) &\equiv 8 \int \frac{d\omega' d\omega''}{(2\pi)^2} G_{n'd}(\omega') G_{b'c}(-\omega') G_{m'a'}(-\omega' + \omega) G_{qn}(\omega'') G_{bm}(-\omega'' + \omega) G_{pa}(-\omega'') \bar{\Gamma}_{mnm'n'}^{(b)}(\omega) \\ &\times \mathcal{L}_{m'_j n'_j a' b'} \mathcal{L}_{m_i n_i ab} \Delta_{cd} \Delta_{pq}^* + 8 \int \frac{d\omega' d\omega''}{(2\pi)^2} G_{qn}^*(\omega') G_{pa'}^*(-\omega') G_{b'm}^*(-\omega' - \omega) G_{n'd}^*(\omega'') G_{bc}^*(-\omega'') \\ &\times G_{m'a}^*(-\omega'' - \omega) \bar{\Gamma}_{mnm'n'}^{(b)*}(-\omega) \mathcal{L}_{m'_j n'_j a' b'} \mathcal{L}_{m_i n_i ab} \Delta_{cd} \Delta_{pq}^*. \end{aligned} \quad (C13d)$$

Equations (C10), (C11), and (C13), together with Eqs. (C8) and (C9), determine the Green function defined by Eq. (C1c). However, this expression contains a large number of terms. After some tedious transformations the Green function $\mathcal{F}_{m_i n_i m'_j n'_j} + (\omega)$ can be finally written in a compact form

$$\begin{aligned} \mathcal{F}_{m_i n_i m'_j n'_j} + (\omega) &= \Delta_{pq}^* \bar{F}_{pqp'q'}(0) [\mathcal{L}_{m_i n_i p' a'} \delta_{q' b'} + \mathcal{L}_{m_i n_i q' b'} \delta_{p' a'}] [\mathcal{L}_{m'_j n'_j a' c'} \delta_{b d'} + \mathcal{L}_{m'_j n'_j b d'} \delta_{a c'}] \\ &\times \bar{F}_{c' d' c d}(0) \Delta_{cd} [\bar{F}_{a' b' a b}(\omega) + 2 \bar{F}_{a' b' u' v'}(\omega) \bar{\Gamma}_{u' v' uv}^{(b)}(\omega) \bar{F}_{uvab}(\omega)] + \text{c.c.}, \end{aligned} \quad (C14)$$

where c.c. means complex conjugation and change the sign of ω .

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