Bosonized squeezed-state coupled-cluster approach to electron correlations in nonlinear spectroscopy

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The signatures of excited-state correlations in the third-order optical response of many-electron systems are calculated using a time-dependent quasiparticle picture which maps the optical excitations onto a system of interacting bosons, following a transformation to boson (oscillator) variables. The many-body wavefunction is approximated by a Gaussian wavepacket representing a squeezed state in the quasiparticle space. Unlike the standard coupled-cluster technique, the present approach constitutes a generalized coherent state representation which makes it possible to derive simple closed equations of motion for its parameters. The time-dependent Hartree–Fock approximation is recovered for weakly correlated systems where squeezing is negligible. © *1999 American Institute of Physics.* [S0021-9606(99)30734-0]

I. INTRODUCTION

The dynamics of strongly correlated many-electron systems constitutes a rapidly developing area of condensed matter physics which includes high-temperature superconductivity,[1,2] the metal–insulator transition,[3,4] and systems of interacting electrons in a strong magnetic field (e.g., the quantum Hall effect).[5,6] Electron correlations are also very important in determining the structure and photoinduced dynamics of conjugated molecules. The single-electron states of conjugated molecules (π-orbitals) are usually delocalized over the entire molecule. Electron correlations are especially strong in conjugated molecules because of their low (1D) dimensionality. The significance of electron correlations in the optical response of various materials can be estimated using the ratio of the exciton binding energy to the optical band gap. In undoped inorganic semiconductors this ratio is $\sim 10^{-2}-10^{-3}$, whereas in conjugated systems this parameter is of the order of one. This implies that processes which do not conserve the number of electron-hole pairs are important in conjugated molecules, and correlations in the excited states are no less important than in the ground state. An interesting problem which arises from strong electron correlations is the relative energy of the lowest excited states which possess the A_g and B_u symmetry. It has been shown experimentally[7,8] and theoretically[9,10] (by exact diagonalization of the *PPP* Hamiltonian) that in short polyenes the lowest (dark) A_g excited state lies below the lowest (bright) B_u state. The correct relative position of the lowest A_g and B_u transition may not be obtained from low-level treatments of correlations, e.g., single CI (Ref. 11) or the time-dependent Hartree–Frock (TDHF).[12–14] This issue has important implications on the photophysics and photochemistry of many conjugated systems. The fluorescence quantum yield drops substantially when the A_g state lies below the B_u state due to nonradiative quenching. Carotenoids are essential components of photosynthetic antenna complexes.[15–17] The energetics of the A_g and B_u transitions in carotenoids controls the energy transfer pathways between the $B800$ and $B850$ bands of the $LH2$ complex.[18] Another problem which requires an improved treatment of correlations is two-photon spectroscopy of strongly correlated systems of electrons in the magnetic field, e.g., when biexcitons which show up in two-photon resonances are formed by Skyrmionic excitons rather than ordinary magnetoexctions.[19]

Electron correlations in the ground state can be treated using density functional theory (DFT).[20–22] Obtaining relevant information on the excited states using this approach constitutes a more complicated task,[23–25] and a convenient way to obtain this information is to apply the time-dependent density functional theory (TDF) which is based on a functional of the time-dependent density.[26–28] Stationary DFT is based on the expansion of the functional in gradients of the charge density.[29,30] In the time-dependent case such an expansion faces some difficulties since the expansion in time derivatives fails and a Pade-type functional has been introduced in the frequency-domain.[27] This functional, however, turns out to be extremely nonlocal[31] and an extension of the theory to current dependent functionals is needed.[32] The TDHF approach[33] is a dynamical variational technique which provides a simple and partial account of excited state correlation effects. It has been applied to study electronic properties of conjugated polymers and molecules.[12–14,34,35] The relevant information on the excited states is contained in the eigenmodes of the linearized TDHF equation.[36] However, the TDHF fails to predict the correct order of the A_g and B_u states in strongly correlated systems.

In this paper we develop a formalism for computing the optical response of many-electron systems, which extends the TDHF and accounts for electron correlations in the excited states in a more profound way. The approach is based on the quasiparticle picture of the optical response whereby the linear response is formed by exciting the quasiparticles, whereas optical nonlinearities originate from their scattering. The classical quasiparticle (oscillator) picture of optical response has been established earlier[37] where it has been demonstrated that the TDHF approximation is the classical limit of the original quantum fermion model (QFM). Choosing a system of local coordinates in the vicinity of the stationary point $\bar{\rho}$ of the TDHF equation maps the system onto a set of

coupled oscillators (quasiparticles), representing electron-hole excitations. Transforming to canonical variables, i.e., to variables for which the Poisson bracket adopts the canonical form (which is always possible locally), one obtains a classical system of interacting bosons, which represent the quasiparticles. Quantizing the classical quasiparticles one arrives at a quantum system of interacting bosons, which represent the elementary excitations of the original QFM.

Representing a system for which the original elementary variables are nonboson (e.g., fermion or spin systems) in terms of effective bosons is known as bosonization. There are two approaches to bosonization.[33] The first is based on immersing the space of states of the original system into the oscillator space.[38] The second uses an expansion of the original basic operators in terms of boson operators.[39,40] Bosonization schemes have been first developed for spin systems.[38–40] The Holstein–Primakoff (HP) expansion[40] constitutes an interesting example of a bosonization scheme of the second type, which contains a parameter s related to the value of the spin. The HP bosonization therefore allows to treat any value of the spin within the same scheme by choosing the proper value of the parameter. For large values of s, the HP bosonization can be considered as the transformation to canonical variables in the phase space of the classical spin represented by the 2D-sphere S^2.[41] Introducing the artificial ''Planck constant'' $\hbar \equiv (2s)^{-1}$, the HP bosonization can be considered as the extension of the canonical transformation to the quantum case. The classical canonical transformation reproduces the HP transformation to first-order in $\hbar$. Extension to the quantum case can then be performed order-by-order in $\hbar$, which constitutes a semiclassical expansion of the HP transformation. The approach developed in this paper extends the HP bosonization from spins to many-electron systems and develops a quantum oscillator (boson) model (QOM) to lowest-order in the artificial Planck constant $\hbar$. This extension is based on incorporating the QFM whose space of states forms an irreducible representation of the unitary group $U(M)$ where M is the number of single-electron states (see Sec. II for details of the representation theory analysis) into a family of irreducible representations of $U(M)$ parametrized by an integer m. The representations of this family are uniquely characterized by the property that they can be obtained using the procedure of geometrical quantization on an orbit given by the Grassmanian manifold $G(N,M;C)$ (see Sec. II for the definition of a Grassmanian),[41] where N is the number of electrons. The case $m=1$ corresponds to the QFM and the artificial Planck constant is defined as $\hbar \equiv m^{-1}$. The case $N=1$, $M=2$ corresponds to the spin, since $G(1,2;C) \cong S^2$ and $m=2s$. Solution of the time evolution of the QOM model, which properly takes into account interactions between the quasiparticles, is not straightforward and requires further approximations. The boson nature of the QOM allows to apply the dynamical variational approach using some natural ansatz for the wavefunctions. The coherent state ansatz for the bosons brings us back to the TDHF approximations. The squeezed state is a natural extension of the coherent state ansatz for a boson system, which is extensively used in the description of quantum states of the radiation field,[42] quantum teleportation,[43] and Bose condensation.[44] It will be shown in Sec. II that the squeezed state ansatz can be considered a bosonized version of the coupled-cluster (CC) ansatz for many-electron systems.[45,46] The latter constitutes an extension of the HF ansatz, includes ground state correlations, and in its dynamical form[47,48] provides relevant information on the excited states. Technically it is much more convenient to work with the boson than the fermion CC ansatz, since the linear and bilinear combinations of the boson operators form a closed algebra and the squeezed states constitute generalized coherent states in the sense of Perelomov.[49,50]

The existence of a closed algebra simplifies the procedure of deriving the variational equations of motion. Let a set of operators $\hat{A}_j$ form a closed algebra: $[\hat{A}_i,\hat{A}_j]=\Sigma f_{ij,k}\hat{A}_k$. We can implement an ansatz for the wavefunctions in the form $\exp(\Sigma_j a_j \hat{A}_j)|\Psi_0\rangle$ where $|\Psi_0\rangle$ constitutes an arbitrary reference state whereas the set of numbers a_j parametrizes the wavefunction. The variational equations of motion are derived from the minimal action principle, with the action $S[\Omega(\tau)]$:

$$S[\Omega(\tau)]=\int dt[i<\Omega(\tau)|d\Omega(\tau)/d\tau>-\langle\Omega(\tau)|\hat{H}|\Omega(\tau)\rangle]. \tag{1.1}$$

Here $\hat{H}$ is the Hamiltonian and $|\Omega(\tau)\rangle$ the time-dependent wavefunction and, therefore, constitute equations of motion of classical dynamics. These equations can be written in the Hamiltonian form for any set B_j of coordinates, which parametrize $|\Omega\rangle$:

$$\frac{dB_j}{d\tau}=\{H,B_j\}. \tag{1.2}$$

The classical Hamiltonian H is given by:

$$H(\Omega)=\langle\Omega|\hat{H}|\Omega\rangle. \tag{1.3}$$

The Poisson bracket adopts a very simple form if the wavefunctions are parametrized by the expectation values $A_j \equiv \langle\Omega|\hat{A}_j|\Omega\rangle$ of the operators $\hat{A}_j$ rather than the set a_j of the ansatz:

$$\{A_m,A_n\}=i\sum_k f_{mn,k}A_k. \tag{1.4}$$

In particular, if the corresponding classical Hamiltonian contains only finite-order terms in the expansion in A_j:

$$H=\sum_{n=1}^{k}\sum_{i_1\ldots i_n} h^{(n)}_{i_1\ldots i_n}A_{i_1}\cdots A_{i_n}, \tag{1.5}$$

then the equations of motion for A_m will also be truncated at the same order. These equations which can be derived from Eq. (1.2) by making use of Eqs. (1.4), (1.5), and the differential property of the Poisson bracket [Eq. (3.8)], have the form:

$$i\frac{dA_m}{d\tau}=\sum_{n=1}^{k}\sum_{j=1}^{n}\sum_{i_0 i_1\cdots i_n} f_{mi_0,\ldots,i_j}h^{(n)}_{i_1\cdots j-1 i_0 i_{j+1}\cdots i_n}A_{i_1}\cdots A_{i_n}. \tag{1.6}$$

This implies that the existence of a closed algebra yields simple equations in terms of the observables. Since, usually, the bottleneck of the dynamical variational approach is relating the parameters of the ansatz to the observables, the problem is simplified considerably if the operator algebra is closed, which is the case of the boson CC ansatz. However, since it is applied to the QOM which constitutes an approximation for the QFM, it still constitutes an approximation for the ordinary fermion CC approach.

The present approach establishes the quasiparticle picture of the optical response of many-electron system. The boson coupled cluster model (BCCM), which is obtained from the QOM by implementing the squeezed state ansatz, can be formulated as classical dynamics in the extended phase space of one- and two-exciton variables. When processes which do not conserve the number of excitons can be neglected (e.g., for Frenkel-excitons), the corresponding equations of motion are exact up to the third-order response. The two-exciton variables give rise to new resonances which are completely neglected within the TDHF approach. Interaction between these two types of variables in the BCCM leads to the decrease of the frequencies of the one-exciton resonances, which has a potential of resolving the A_g, B_u problem within a relatively simple calculation. It is possible to avoid the bosonization altogether and derive directly equations of motion for the expectation values of the bilinear and fourth-order combinations of the fermion operators.[51] However, the present scheme has several advantages. First, it has precisely the right number of variables (we have one quasiparticle per an electron-hole pair), whereas when working with Fermion objects we have redundant variables due to the over-completeness of the basis set. This allows to avoid the difficulties which arise from including the intraband variables.[51] All the necessary sum rules are therefore automatically satisfied in the present approach. Second, it established an oscillator picture of optical response. Finally, our approach can be easily extended along the lines of Ref. 52 to treat incoherent dynamical processes, which arise from the coupling to nuclear motions.

The resulting computational scheme for the optical response consists of the four steps illustrated in Fig. 1: (i) We start with the original many-electron quantum fermion model (QFM), and introduce its classical counterpart. The phase space of the corresponding classical dynamics related to the TDHF (coherent state) approximation is the space M of single Slater determinants. Following,[37] the classical Hamiltonian is defined by Eq. (1.3) and constitutes a function on M. The Poisson bracket in M has been introduced in Ref. 37. This yields the classical oscillator model (COM). (ii) We introduce a reference Slater determinant Ω_0 which is the solution of the stationary HF equation, and find the canonical variables in the vicinity of Ω_0. The system described in terms of the canonical variables constitutes a system of classical coupled oscillators, hereafter referred to as the classical canonical oscillator model (CCOM). (iii) The CCOM is quantized to arrive at the quantum oscillator model (QOM) which represents a quantum system of interacting boson quasiparticles. (iv) The squeezed state ansatz is applied to the quantum oscillator, yielding the boson coupled cluster model (BCCM). The resulting variational equations of motion provide a dynamical theory for the optical response which extends the TDHF by incorporating additional two-exciton dynamical variables.

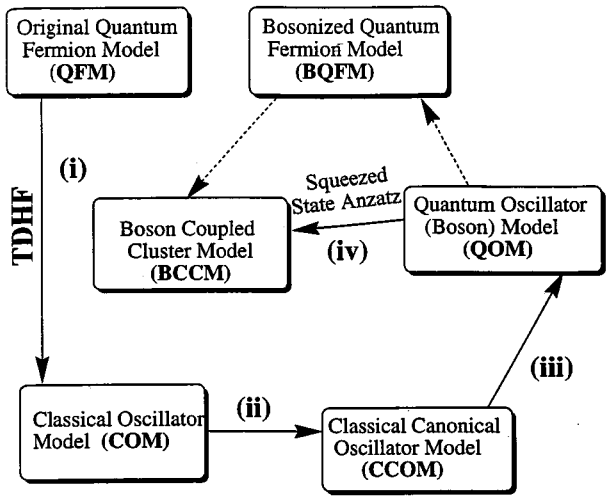

FIG. 1. The solid arrows illustrate the four steps involved in constructing the boson coupled cluster classical model (BCCM) out of the original quantum fermion model (QFM). Obtaining a bosonized quantum fermion model (BQFM) out of the quantum oscillator model, which can be carried out order-by-order in the artificial Planck constant, should allow to incorporate higher-order corrections in the strength of electron correlations. This extension (dashed arrows) which follows the generalization of the Holstein–Primakoff bosonization scheme (Ref. 40) to the case of many-electron systems goes beyond the scope of the present work.

This paper is organized as follows. In Sec. II we introduce the boson coupled-cluster expansion based on the squeezed states and compare it with the regular fermion CC expansion. Section III is devoted to developing a boson representation of the fermion system [steps (i), (ii), and (iii)]. In Sec. IV we apply the squeezed state ansatz and derive closed equations of motion for its parameters [Step (iv)]. Our results are finally discussed in Sec. V. For clarity, details of most derivations are moved to the Appendices, when possible.

II. FERMION VS. BOSON COUPLED-CLUSTER EXPANSIONS

We start with the general Hamiltonian describing an optically driven many-electron system with two-body interactions.

$$\hat{H}(\tau)=\sum\ t_{mn}\hat{C}_m^+\hat{C}_n+\frac{1}{2}\sum\ V_{mn,ke}\hat{C}_m^+\hat{C}_n^+\hat{C}_k\hat{C}_e-\varepsilon(\tau)\hat{P}. \tag{2.1}$$

Here $\hat{C}_m(\hat{C}_m^+)$ are the electron annihilation (creation) operators, with the commutation relations

$$\hat{C}_m\hat{C}_n^+ + \hat{C}_n^+\hat{C}_m=\delta_{mn}, \tag{2.2}$$

and the Latin indices label the single-electron states (including spin). $\varepsilon(\tau)$ is a classical driving field and the polarization operator has the form of a general single-electron operator

$$\hat{P}=\sum\ \mu_{mn}\hat{C}_m^+\hat{C}_n. \tag{2.3}$$

A dynamical variational approach, based on an assumption that the many-body wavefunction of the driven system belongs at all times to a certain manifold M, constitutes a useful tool for building approximate methods for the dynamics of the driven system. It further provides reduced relevant information on the eigenstates obtained by investigating the resonant structure of the frequency-domain optical response. A variational scheme is fully determined by selecting the manifold M to which the system dynamics is restricted. The choice of M also constitutes an approximation for the ground state which is obtained by minimizing the system's energy on M (the standard variational procedure). The approximation for the ground state must be compatible with the dynamics: The ground state is given by a stationary point of the dynamical equations in the absence of the driving field.

By choosing M to be the space of all single Slater determinants, one obtains the Hartree–Fock (HF) approximation for the ground state and the time-dependent Hartree–Fock (TDHF) approximation for the dynamics. Both HF and TDHF are extremely powerful, however they neglect electron correlations in the ground state, and are restricted to two-particle correlations in the excited states. The space of single Slater determinants constitutes a homogeneous manifold. In the case of N electrons which can occupy M single-electron orbitals ($N<M$), it is represented by the Grassman manifold $G(N,M;C)$ of N-dimensional vector subspaces of the M-dimensional vector space C^M. $G(N,M;C)$ can be represented as a factor space $G(N,M;C)\cong U(M)/U(N)\times U(M-N)$ with the natural action of $U(M)$ on it, where $U(n)$ denotes the group of $n\times n$ unitary matrices. The action of $U(M)$ on $G(N,M;C)$ is induced by its natural action on the orbitals representing the Slater determinants. All possible Slater determinants can be generated from a single one by acting with the elements of $U(M)$. The Lie algebra corresponding to $U(M)$ is generated by bilinear combinations of the creation and annihilation operators $\hat{C}_m^+ C_n$ which form a closed algebra with respect to the commutator:

$$[\hat{C}_m^+\hat{C}_n,\hat{C}_{m'}^+,\hat{C}_{n'}]=\delta_{nm'}\hat{C}_m^+\hat{C}_{n'}-\delta_{n'm}\hat{C}_{m'}^+,\hat{C}_n. \tag{2.4}$$

Choosing a reference Slater determinant $|\Omega_0\rangle$ we have $|\Omega(\tau)\rangle=\mathscr{g}|\Omega_0\rangle$ for some $\mathscr{g}\in U(M)$ which implies that single Slater determinants constitute a set of generalized coherent states in the sense of Perelomov for a unitary group.[53] Applying an exponential representation for $\mathscr{g}\in U(M)$ in terms of an element of the corresponding Lie algebra $\mathscr{g}=\exp(\Sigma U_{mn}\hat{C}_m^+\hat{C}_n)$ yields the Thouless representation of a Slater determinant.[33]

$$|\Omega(\tau)\rangle=\exp\left(\sum U_{mn}(\tau)\hat{C}_m^+\hat{C}_n\right)|\Omega_0\rangle. \tag{2.5}$$

Note that even though the sum in Eq. (2.5) runs over all single-electron states, we can make a transformation to the natural orbitals (eigenvectors of the density matrix) and in the sum $\Sigma_{\alpha\beta}U_{\alpha\beta}\hat{C}_\alpha^+\hat{C}_\beta$ only retain terms where α represents unoccupied orbitals (i.e., $\hat{C}_\alpha|\Omega_0\rangle=0$) and β stands for occupied orbitals (i.e., $\hat{C}_\beta^+|\Omega_0\rangle=0$). Although the elements $\exp(\Sigma_{\alpha\beta}U_{\alpha\beta}\hat{C}_\alpha^+\hat{C}_\beta)$ with this restricted summation do not represent all elements $\mathscr{g}$ of the group $U(M)$ even in the vicinity of the unit element of the group, $\exp(\Sigma_{\alpha\beta}U_{\alpha\beta}\hat{C}_\alpha^+\hat{C}_\beta)|\Omega_0\rangle$ still represents all coherent states (at least in the vicinity of $|\Omega_0\rangle$). This is the case since the elements $\exp[\Sigma_{\alpha\alpha'}(U_{\alpha\alpha'}\hat{C}_\alpha^+\hat{C}_{\alpha'}+\Sigma_{\beta\beta'}U_{\beta\beta'}\hat{C}_\beta^+\hat{C}_{\beta'})]$, where α,α' are unoccupied, and β,β' are occupied orbitals, do not affect $|\Omega_0\rangle$. TDHF equations of motion for the coupled electronic and nuclear dynamics have been derived using a generalized coherent state as a trial wavefunction.[54]

A straightforward generalization of the TDHF scheme is obtained by extending the ansatz for the wavefunctions to include new parameters. The coupled cluster approach is a class of ansatzes which include the single Slater determinants as special cases.[45,46] This ansatz, widely used to treat ground state correlations, is obtained by adding the fourth-order terms to the Thouless representation:

$$|\Omega(\tau)\rangle=\exp\left(\sum U_{mn}(\tau)\hat{C}_m^+\hat{C}_n+\sum V_{mn,ke}(\tau)\hat{C}_m^+\hat{C}_n^+\hat{C}_k\hat{C}_e\right)|\Omega_0\rangle. \tag{2.6}$$

Closing the dynamical equations for $U_{mn}(\tau)$ and $V_{mn,ke}(\tau)$ is a formidable task, and further approximations need to be introduced. The difficulties can be rationalized using the theory of representations. The bilinear and fourth-order combinations of fermion operators do not form a closed algebra, since commutators of the fourth-order terms produce sixth-order terms, etc. This implies that the coupled-cluster (unlike the HF) states may not be considered generalized coherent states.

The algebra of electron-hole operators $\hat{C}_m^+\hat{C}_n$ may not generally be extended to a closed algebra of a reasonable dimensionality, which implies that the TDHF Ansatz does not have extensions that have properties of generalized coherent states. Such extensions, however, do exist if the basic operators are bosons. Consider a system of coupled oscillators and let $B_m(B_m^+)$ be a set of annihilation (creation) boson operators, which form the Heisengberg–Weyl algebra:

$$[\hat{B}_m,\hat{B}_n^+]=\delta_{mn}\hat{I}. \tag{2.7}$$

The generalized coherent states related to the Heisenberg–Weyl group, which correspond to the algebra of Eq. (2.7) are the ordinary coherent states:

$$|\Omega(\tau)\rangle=\exp\left(\sum_m b_m(\tau)B_m^+\right)|\Omega_0\rangle, \tag{2.8}$$

where $|\Omega_0\rangle$ is the ground state. An extended Heisenberg–Weyl algebra can be obtained by adding bilinear combinations of the boson operators. We introduce the operators

$$\hat{X}^{(-)}_{mn} \equiv \hat{B}_m \hat{B}_n, \quad \hat{X}^{(+)}_{mn} \equiv \hat{B}^+_m \hat{B}^+_n, \quad \hat{X}^{(z)}_{mn} \equiv \hat{B}^+_m \hat{B}_n + \tfrac{1}{2}\delta_{mn}\hat{I}, \tag{2.9}$$

which satisfy the following commutation relations.

$$\begin{aligned}
&[\hat{X}^{(-)}_{mn,}\hat{X}^{(+)}_{rs}] = \delta_{nr}\hat{X}^{(z)}_{sm} + \delta_{ns}\hat{X}^{(z)}_{rm} + \delta_{ms}\hat{X}^{(z)}_{rn} + \delta_{mr}\hat{X}^{(z)}_{sn}, \\
&[\hat{X}^{(z)}_{mn,}\hat{X}^{(-)}_{rs}] = -\delta_{mr}\hat{X}^{(-)}_{ns} - \delta_{ms}\hat{X}^{(-)}_{nr}, \\
&[\hat{X}^{(z)}_{mn,}\hat{X}^{(+)}_{rs}] = \delta_{nr}\hat{X}^{(+)}_{ms} + \delta_{ns}\hat{X}^{(+)}_{ms}.
\end{aligned} \tag{2.10}$$

The mixed commutators between the bilinear and linear operators have the form:

$$\begin{aligned}
&[\hat{X}^{(+)}_{mn,}\hat{B}_j] = -\delta_{mj}\hat{B}^+_n - \delta_{nj}\hat{B}^+_m; \;\; B^+_{m,}; \;\; [\hat{X}^{(z)}_{mn,}\hat{B}_j] = -\delta_{mj}\hat{B}_n, \\
&[\hat{X}^{(-)}_{mn,}\hat{B}^+_j] = \delta_{nj}\hat{B}_m + \delta_{mj}\hat{B}_n; \;\; B^+_{n,}; \;\; [\hat{X}^{(z)}_{mn,}\hat{B}^+_j] = \delta_{nj}\hat{B}^+_m.
\end{aligned} \tag{2.11}$$

All other commutators vanish.

The generalized coherent states with respect to the extended Heisenberg–Weyl algebra (group) are the squeezed states.[49]

$$|\psi(\tau)\rangle = \exp\left(\sum_m b_m(\tau)\hat{B}^+_m + \sum_{mm} c_{mn}(\tau)\hat{B}^+_n\hat{B}^+_m\right)|\Omega_0\rangle. \tag{2.12}$$

As shown in Eq. (1.6), for variational applications it is more convenient to parametrize these states by the set of expectation values of linear and binary operators (instead of b_m and c_{mn}):

$$B_m \equiv \langle\psi|\hat{B}_m|\psi\rangle, \quad Y_{mn} \equiv \langle\psi|\hat{B}_m\hat{B}_n|\psi\rangle - B_mB_n, \tag{2.13}$$

$$N_{mn} \equiv \langle\psi|\hat{B}^+_m\hat{B}_n|\psi\rangle - B^*_mB_n. \tag{2.14}$$

We further have

$$B^*_m = \langle\psi|\hat{B}^+_m|\psi\rangle, \quad Y^*_{mn} = \langle\psi|\hat{B}^+_m\hat{B}^+_n|\psi\rangle - B^*_mB^*_n. \tag{2.15}$$

Using the squeezed state ansatz, the variables N and Y are not independent and N can be eliminated (see Appendix A)

$$N = \sqrt{\frac{1}{4}I + Y^+Y} - \frac{1}{2}I. \tag{2.16}$$

Here N and Y are linear operators with matrix elements N_{mn} and Y_{mn}, and I is the unit operator: $I_{mn} = \delta_{mn}$. B_m and Y_{mn} constitute a very convenient parametrization of the squeezed states. B_m represents the average position of the state, whereas Y_{mn} are responsible for the squeezing. An ordinary (nonsequeezed) coherent state has $Y_{mn} = 0$. The expectation value of any operator (such as the Hamiltonian) expanded as a normally ordered product of the form $\hat{B}^+_{i_1}\cdots\hat{B}^+_{i_m}\hat{B}_{j_1}\cdots\hat{B}_{j_n}$ can be expressed in terms of B, Y, and N by applying the Wick theorem together with Eq. (2.16):

$$N = Y^+Y - (Y^+Y)^2 + 4(Y^+Y)^3 - \ldots. \tag{2.17}$$

Let us now return to the problem of extending the TDHF for the many-electron system, and compare the difficulties with those for a system of oscillators. Both systems can be described in terms of generalized coherent states (Slater determinants for the electrons and ordinary coherent states for the oscillators). An attempt to go beyond the coherent state ansatz by considering wavepackets of coherent states of a form $\int d\Omega f(\Omega)|\Omega\rangle$ faces some difficulties in both cases due to the over-completeness of the coherent states. However, in contrast to the fermion system, the boson system has another representation in terms of functions of the oscillator coordinates alone (we reiterate that a wavepacket $\int d\Omega f(\Omega)|\Omega\rangle$ is determined by a function $f(\Omega)$ in phase space, i.e., coordinates and momenta) which form a complete basis set. This allows to extend the coherent state ansatz for the oscillator system using the coordinate representation. The two systems also behave differently under an attempt to extend the coherent state ansatz algebraically. The algebra of the boson operators has a natural extension to a closed algebra achieved by adding the bilinear combinations of the basic operators. This leads to the squeezed state ansatz. A similar procedure for the electron system leads to the coupled cluster ansatz, where the corresponding extended algebra is not closed.

The difficulties connected with the original fermion system can be avoided by mapping it onto an effective system of oscillators. Formally this can be accomplished by representing the bilinear combinations of the fermion operators in terms of effective bosons, i.e., we expand $C^+_mC_n$ as an infinite power series of $\hat{B}$ and $\hat{B}^+$. Such a bosonization scheme can be constructed by making use of a fact that a transformation to canonical dynamical variables is a classical counterpart of the bosonization. This will be done in the next section.

III. COHERENT-STATE-BASED CLASSICAL BOSONIZATION OF THE TDHF

In this section we derive the classical limit of the QFM and recast it in terms of classical oscillators. This will accomplish steps (i) and (ii) of Fig. 1. Step (iii) is then carried out in Appendix D.

Following Ref. 37 we introduce the space of single Slater determinants, $M = G(N,M;C)$ which is the Grassman manifold with the natural action of the group $U(M)$, as described in Sec. II. The classical Hamiltonian can be conveniently expressed in terms of a set ρ_{mn} of functions on M defined by

$$\rho_{mn}(\Omega) \equiv \langle\Omega|\hat{C}^+_m\hat{C}_n|\Omega\rangle. \tag{3.1}$$

$\rho_{mn}(\Omega)$ are the components of the single-electron reduced density matrix corresponding to the single Slater determinant $|\Omega\rangle$. The classical Hamiltonian is easily obtained upon the substitutions of Eq. (2.1) into Eq. (1.3) and making use of Eq. (3.1) and the Wick theorem for single Slater determinants:

$$H(\tau) = \sum t_{mn}\rho_{mn} + \frac{1}{2}\sum V_{mn,ke}(\rho_{me}\rho_{nk} - \rho_{mk}\rho_{ne}) - \varepsilon(\tau)P, \tag{3.2}$$

with

$$P = \sum \mu_{mn}\rho_{mn}. \tag{3.3}$$

Deriving a classical equation of motion for ρ_{mn} requires the introduction of a Poisson bracket. The Poisson bracket on M may be defined as follows: We first introduce a differen-

tial one-form a on M by $a \equiv \langle \Omega | d\Omega \rangle$, which gives rise to the closed 2-form $\omega \equiv da$, ω defines a simplectic structure on M. Note that the state $|\Omega\rangle$ is defined up to a phase. Although the form a depends on the phase, since

$$a' = \langle e^{i\varphi(\Omega)}\Omega | d[e^{i\varphi(\Omega)}\Omega] \rangle = a + i d\varphi, \tag{3.4}$$

ω does not depend on that phase since $da' = da$. The Poisson bracket is the dual object to the simplectic structure ω, so that in a system of local coordinates

$$\omega = \sum_{ik} \omega^{ik}(x) dx_i \wedge dx_k, \tag{3.5}$$

the Poisson bracket of two functions f and g adopts a form

$$\{f,g\} = \sum_{ik} \omega_{ik}(x) \frac{\partial f}{\partial x_i} \frac{\partial g}{\partial x_k}, \tag{3.6}$$

where $\Sigma_k \omega_{ik}(x)\omega^{kj}(x) = \delta_{ij}$. This procedure is based on the dynamical variational principle which defines a classical Lagrangian mechanics on M. It reflects the standard derivation of the Hamiltonian classical dynamics formulation from the Lagrangian.

Equation (3.6) defines the Poisson bracket in a differential form, i.e., geometrically. Since M is a space of generalized coherent states with respect to the group $U(M)$, the same Poisson bracket can be introduced algebraically,[37] and we have

$$\{\rho_{mn,}\rho_{kl}\} = i(\delta_{nk}\rho_{ml} - \delta_{lm}\rho_{kn}). \tag{3.7}$$

The Poisson bracket can be extended to any functions of ρ by making use of the differential property

$$\{f,gh\} = -\{gh,f\} = \{f,g\}h + g\{f,h\}. \tag{3.8}$$

The resulting classical equation of motion, $\dot{\rho} = \{H(\tau),\rho\}$, finally yields the TDHF equation

$$i\frac{d\rho}{d\tau} = [t + V(\rho),\rho], \tag{3.9}$$

where $V(\rho)_{mn} \equiv -2\Sigma_{m'n'} V_{mn',nn'}\rho_{m'n'}$. This accomplishes step (i) where the TDHF is written as the equations of motion of a classical oscillator model.

Recasting these equations using canonical variables, [step (ii)] will be carried out in two stages. We first introduce almost canonical variables obtained as eigenmode of the linearized TDHF equations and then perform a nonlinear transformation, order by order, to obtain the desired canonical variables. We start by expanding the density matrix in the vicinity of a reference coherent state $|\Omega_0\rangle$:[37]

$$\rho(\tau) = \overline{\rho} + \xi(\tau) + T[\xi(\tau)], \tag{3.10}$$

where ξ and $T(\xi)$ are the interband (particle–hole) and intraband (particle–particle and hole–hole) components of $(\rho - \overline{\rho})$ with respect to $\overline{\rho}$, i.e.,[37] $[T(\xi),\overline{\rho}] = 0$, $[\overline{\rho},[\overline{\rho},\xi]] = 0$. $T(\xi)$ is uniquely determined by ξ, since ρ is idempotent $\rho^2 = \rho$. To second-order in ξ we have

$$T(\xi) = \tfrac{1}{2}[[\xi,\overline{\rho}],\xi] + \cdots. \tag{3.11}$$

We choose $|\Omega_0\rangle$ to be the solution of the stationary HF equation.

$$[t + V(\overline{\rho}),\overline{\rho}] = 0, \tag{3.12}$$

and introduce a system of local coordinates on M in the vicinity of $\overline{\rho}$ by expanding ξ in the form

$$\xi(\tau) = \sum_{\alpha>0} [(z'_\alpha(\tau)\xi_\alpha + z'^*_\alpha(\tau)\xi^+_\alpha)]. \tag{3.13}$$

Here z'_α are expansion coefficients and ξ_α are the eigenmodes of the superoperator L of the linearized TDHF equation [Eq. (B7)]:

$$L\xi_\alpha = \Omega_\alpha \xi_\alpha. \tag{3.14}$$

These eigenmodes come in pairs: for any eigenmode ξ_α, with eigenvalue Ω_α, ξ^+_α is an eigenmode with eigenvalue $-\Omega_\alpha$. $\alpha > 0$ means that $\Omega_\alpha > 0$. We will also use the notation $\xi_{-\alpha} = \xi^+_\alpha$, $z'_{-\alpha} = z'^*_\alpha$. Substituting the expansion of Eq. (3.13) into Eq. (3.10) and making use of Eq. (3.11), we can expand any phase space function in terms of the local coordinates z'_α. In particular, substituting Eq. (3.10) into Eq. (3.2), we obtain the classical Hamiltonian. To fourth-order in z' it reads:

$$H(\tau) = \sum_{\alpha>0} \Omega_\alpha z'^*_\alpha z'_\alpha + \sum_{\alpha\beta\gamma} \overline{V}^{(3)}_{\alpha\beta\gamma} z'_\alpha z'_\beta z'_\gamma + \sum_{\alpha\beta\gamma\delta} \overline{V}^{(4)}_{\alpha\beta\gamma\delta} z'_\alpha z'_\beta z'_\gamma z'_\delta - \varepsilon(\tau)P, \tag{3.15}$$

where

$$P = \sum_\alpha \mu^{(1)}_\alpha z'_\alpha + \frac{1}{2}\sum_{\alpha\beta} \mu^{(2)}_{\alpha\beta} z'_\alpha z'_\beta. \tag{3.16}$$

Note that the summations in the second and the third terms (unlike the first) run over negative as well as positive Greek indices. Closed expressions for all coefficients $\mu^{(1)}$, $\mu^{(2)}$, $\overline{V}^{(3)}$, and $\overline{V}^{(4)}$ in terms of the original Hamiltonian parameters, $\overline{\rho}_{mn}$, and the eigenmodes $\xi_{\alpha,mn}$, are given in Appendix B.

The Poisson bracket of z' can be obtained by starting with Eq. (3.7) and making use of Eqs. (3.10), (3.11), and (3.14). To third-order we obtain:

$$\{z'_\alpha, z'_\beta\} = i\,\mathrm{sgn}(\alpha)\delta_{\alpha-\beta} + i\sum_{\gamma\delta} F_{\alpha\beta,\gamma\delta} z'_\gamma z'_\delta, \tag{3.17}$$

where $F^{(4)}$ is given by Eq. (C9). To first-order, Eq. (3.17) reads $\{z'_\alpha, z'^*_\beta\} = i\delta_{\alpha\beta}$ which corresponds to the canonical commutation relations written in the holomorphic representation.

The dynamics of z'_α is determined by the Liouville equation $\dot{z}'_\alpha = \{H(\tau), z'_\alpha\}$. Using these trajectories we can compute the single electron density matrix

$$\rho(\tau) = \overline{\rho} + \sum_\alpha d^{(1)}_\alpha z'_\alpha(\tau) + \frac{1}{2}\sum_{\alpha\beta} d^{(2)}_{\alpha\beta} z'_\alpha(\tau) z'_\beta(\tau). \tag{3.18}$$

$d^{(1)}_\alpha$ and $d^{(2)}_{\alpha\beta}$ are matrices in the single electron space with matrix elements $(d^{(1)}_\alpha)_{mn}$ and $(d^{(2)}_{\alpha\beta})_{mn}$ (note that α and $\alpha\beta$

are labels and not indices). These matrices can be computed by starting with a matrix d_{mn} representing $\hat{C}_m^+ \hat{C}_n$ in the single electron space (all matrix elements are zero except for the mn element which is equal to 1). The matrices $d_\alpha^{(1)}$ and $d_{\alpha\beta}^{(2)}$ are obtained from Eqs. (B15) by substituting d for μ.

Using $\rho(\tau)$ we can compute the expectation value of any single electron operator such as P

$$P(\tau)=\sum_{mn} \mu_{mn}\rho_{mn}(\tau). \tag{3.19}$$

In particular, this results in Eq. (3.16).

We next transform the variable z'_α to a new set z_α for which the Poisson bracket is canonical:

$$\{z_\alpha, z^*_\beta\}=i\delta_{\alpha\beta}, \tag{3.20}$$

This transformation can be carried out order by order, as shown in Appendix C. To third-order we have:

$$z'_\alpha = z_\alpha + \sum_{\beta\gamma\delta} A^{(3)}_{\alpha,\beta\gamma\delta} z_\beta z_\gamma z_\delta, \tag{3.21}$$

where $A^{(3)}$ is given in Eq. (B11). Substituting Eq. (3.21) into Eq. (3.15) we finally get

$$\begin{aligned} H(\tau)=&\sum_\alpha \Omega_\alpha z^*_\alpha z_\alpha + \sum_{\alpha\beta\gamma} V^{(3,0)}_{\alpha\beta\gamma} z^*_\alpha z^*_\beta z^*_\gamma + \sum_{\alpha\beta\gamma} V^{(2,1)}_{\alpha\beta,\gamma} z^*_\alpha z^*_\beta z_\gamma + \sum_{\alpha\beta\gamma} V^{(1,2)}_{\alpha,\beta\gamma} z^*_\alpha z_\beta z_\gamma \\ &+ \sum_{\alpha\beta\gamma} V^{(0,3)}_{\alpha\beta\gamma} z_\alpha z_\beta z_\gamma + \sum_{\alpha\beta\gamma\delta} V^{(4,0)}_{\alpha\beta\gamma\delta} z^*_\alpha z^*_\beta z^*_\gamma z^*_\delta + \sum_{\alpha\beta\gamma\delta} V^{(0,3)}_{\alpha\beta\gamma,\delta} z^*_\alpha z^*_\beta z^*_\gamma z_\delta + \sum_{\alpha\beta\gamma\delta} V^{(2,2)}_{\alpha\beta,\gamma\delta} z^*_\alpha z^*_\beta z_\gamma z_\delta \\ &+ \sum_{\alpha\beta\gamma\delta} V^{(1,3)}_{\alpha,\beta\gamma\delta} z^*_\alpha z_\beta z_\gamma z_\delta + \sum_{\alpha\beta\gamma\delta} V^{(0,4)}_{\alpha\beta\gamma\delta} z_\alpha z_\beta z_\gamma z_\delta - \varepsilon(\tau) P \end{aligned} \tag{3.22}$$

where the summations are restricted to positive values of the Greek indices. This Hamiltonian constitutes a coupled oscillator representation for the classical counterpart of the fermion system. The parameters $V^{(i,j)}$ are given in Appendix B. Equation (3.22) together with (3.20), (3.18), and (3.21) accomplish step (ii). Step (iii) is now straightforward and is carried out in Appendix D. The classical oscillator Hamiltonian [Eq. (3.22)] is quantized and the resulting quantum oscillator Hamiltonian is given in Eq. (D2). In the classical limit the equations of motion derived from this Hamiltonian coincide with the TDHF equations. However, more generally it constitutes a key step in the generalization of the TDHF, which will be presented next.

IV. SQUEEZED-STATE ANSATZ AND GENERALIZED COHERENT STATE EXTENSION OF THE TDHF

In Appendix E we carry out step (iv) by applying the squeezed-state ansatz for the time-dependent wavefunction of the quantum Hamiltonian [Eq. (D2)]. This yields a new classical Hamiltonian given by Eqs. (E1)–(E4) in the extended phase space of B_α and $Y_{\mu\nu}$ variables. Since the squeezed states form a set of generalized coherent states of the extended Heisenberg–Weyl algebra, and the B, Y, Y^*, and N variables are expectation values of its generators on the squeezed states, their Poisson bracket is given by the commutators of the underlying generators. This yields

$$\begin{aligned} &\{B_\alpha, B^*_\beta\}=i\delta_{\alpha\beta}, \\ &\{Y_{\alpha\beta}, Y^*_{\gamma\delta}\}=i(\delta_{\beta\gamma}\delta_{\alpha\delta}+\delta_{\beta\delta}\delta_{\alpha\gamma})+i(\delta_{\beta\gamma}N_{\delta\alpha}+\delta_{\beta\delta}N_{\gamma\alpha} \\ &\qquad +\delta_{\alpha\gamma}N_{\delta\beta}+\delta_{\alpha\delta}N_{\gamma\beta}), \\ &\{N_{\alpha\beta}, Y_{\gamma\delta}\}=-i(\delta_{\alpha\gamma}Y_{\beta\delta}+\delta_{\alpha\delta}Y_{\beta\gamma}), \\ &\{N_{\alpha\beta}, Y^*_{\gamma\delta}\}=i(\delta_{\beta\gamma}Y^*_{\alpha\delta}+\delta_{\beta\delta}Y^*_{\alpha\gamma}). \end{aligned} \tag{4.1}$$

The equations of motion for the variables B and Y assume the form of the classical Liouville equation:

$$\frac{df}{d\tau}=\{H,f\}, \tag{4.2}$$

where H is given by Eqs. (E1)–(E4) and $f=B_\alpha, Y_{\beta\gamma}$. Since $N_{\alpha\beta}$ are not independent variables and can be expressed in terms of $Y_{\gamma\delta}$ [Eq. (2.16)], these equations can be derived in two ways. The first involves expressing N in terms of Y in the classical Hamiltonian and then using Eq. (4.2). Alternatively we can first apply Eq. (4.2) and then express N in terms of Y in the right hand side of the resulting dynamical equations. In what follows we use the second scheme.

Before deriving the full system of equations of motions we shall first consider the harmonic part of the Hamiltonian and the zero-order Poisson bracket in terms of the B and Y variables, since these determine the coupled eigenmodes of the linearized equation of motion. The Poisson bracket then adopts the canonical form

$$\begin{aligned} &\{B_\alpha, B^*_\beta\}=i\delta_{\alpha\beta}, \\ &\{Y_{\alpha\beta}, Y^*_{\gamma\delta}\}=i(\delta_{\beta\gamma}\delta_{\alpha\delta}+\delta_{\beta\delta}\delta_{\alpha\gamma}), \end{aligned} \tag{4.3}$$

with the harmonic Hamiltonian H_L expressed in terms of B and Y:

$$H_L=H_B+H_Y+H_{BY}, \tag{4.4}$$

where

$$H_B=\sum_\alpha \Omega_\alpha B^*_\alpha B_\alpha, \tag{4.5}$$

is the Hamiltonian of oscillators representing the TDHF modes, and

$$H_Y=\frac{1}{2}\sum_{\alpha\beta}(\Omega_\alpha+\Omega_\beta)Y^*_{\alpha\beta}Y_{\alpha\beta}+\sum_{\alpha\beta\gamma\delta}V^{(2,2)}_{\alpha\beta,\gamma\delta}Y^*_{\alpha\beta}Y_{\gamma\delta}+6\sum_{\alpha\beta\gamma\delta}V^{(4,0)}_{\alpha\beta\gamma\delta}Y^*_{\alpha\beta}Y^*_{\gamma\delta}+6\sum_{\alpha\beta\gamma\delta}V^{(0,4)}_{\alpha\beta\gamma\delta}Y_{\alpha\beta}Y_{\gamma\delta}, \tag{4.6}$$

represents the additional Y-oscillators responsible for the squeezing. Finally

$$H_{BY}=\sum_{\alpha\beta\gamma}V^{(2,1)}_{\alpha\beta,\gamma}Y^*_{\alpha\beta}B_\gamma+\sum_{\alpha\beta\gamma}V^{(1,2)}_{\alpha,\beta\gamma}B^*_\alpha Y_{\beta\gamma}+3\sum_{\alpha\beta\gamma}V^{(3,0)}_{\alpha\beta\gamma}B^*_\alpha Y^*_{\beta\gamma}+3\sum_{\alpha\beta\gamma}V^{(0,3)}_{\alpha\beta\gamma}B_\alpha Y_{\beta\gamma}, \tag{4.7}$$

describes the coupling of the B and Y oscillators. The Y-oscillators can be considered as pairs of B-oscillators, the first term in the r h s of Eq. (4.6) represents the energy of two noninteracting B-oscillators whereas the other correction terms are responsible for the interaction.

To describe the optical response up to third-order we expand the equations of motion, retaining cubic terms in B and Y. We then have:

$$\begin{aligned} i\frac{dB_\alpha}{d\tau}&=\Omega_\alpha B_\alpha+\sum_{\beta\gamma}V^{(1,2)}_{\alpha,\beta\gamma}Y_{\beta\gamma}+3\sum_{\beta\gamma}V^{(3,0)}_{\alpha\beta\gamma}Y^*_{\beta\gamma}+\dot{B}_\alpha,\\ i\frac{dY_{\mu\nu}}{d\tau}&=(\Omega_\mu+\Omega_\nu)Y_{\mu\nu}+2\sum_{\gamma\delta}V^{(2,2)}_{\mu\nu,\gamma\delta}Y_{\gamma\delta}+12\sum_{\alpha\beta}V^{(4,0)}_{\mu\nu\alpha\beta}Y^*_{\alpha\beta}+6\sum_{\alpha}V^{(3,0)}_{\mu\nu\alpha}B^*_\alpha\\ &\quad+2\sum_{\gamma}V^{(2,1)}_{\mu\nu,\gamma}B_\gamma+\dot{Y}_{\mu\nu}, \end{aligned} \tag{4.8}$$

where $\dot{B}$ and $\dot{Y}$ denote the nonlinear terms which are given in Appendix E.

Using the solution of these equations, the density matrix is given by:

$$\begin{aligned} \rho(\tau)&=\overline{\rho}_{mn}+\left[\sum_\alpha d^{(1,0)}_\alpha B^*_\alpha+\text{c.c.}\right]+\frac{1}{2}\left[\sum_{\alpha\beta}d^{(2,0)}_{\alpha\beta}B^*_\alpha(\tau)B^*_\beta(\tau)+\text{c.c.}\right]\\ &\quad+\frac{1}{2}\left[\sum_{\alpha\beta}d^{(2,0)}_{\alpha\beta}Y^*_{\alpha\beta}(\tau)+\text{c.c.}\right]+\sum_{\alpha\beta}d^{(1,1)}_{\alpha,\beta}B^*_\alpha(\tau)B_\beta(\tau)+\sum_{\alpha\beta\gamma}d^{(1,1)}_{\alpha,\beta}Y^*_{\alpha\gamma}(\tau)Y_{\beta\gamma}(\tau) \end{aligned} \tag{4.9}$$

Finally, using Eq. (3.19) and (4.9), the polarization assumes the following form:

$$\begin{aligned} P&=\left[\sum_\alpha\mu^{(1,0)}_\alpha B^*_\alpha+\text{c.c.}\right]+\frac{1}{2}\left[\sum_{\alpha\beta}\mu^{(2,0)}_{\alpha\beta}B^*_\alpha B^*_\beta+\text{c.c.}\right]\\ &\quad+\frac{1}{2}\left[\sum_{\alpha\beta}\mu^{(2,0)}_{\alpha\beta}Y^*_{\alpha\beta}+\text{c.c.}\right]+\sum_{\alpha\beta}\mu^{(1,1)}_{\alpha,\beta}B^*_\alpha B_\beta\\ &\quad+\sum_{\alpha\beta\gamma}\mu^{(1,1)}_{\alpha,\beta}Y^*_{\alpha\gamma}Y_{\beta\gamma}. \end{aligned} \tag{4.10}$$

The TDHF is recovered from these equations by simply setting $Y=0$.

V. DISCUSSION

The present derivation of the oscillator equations of motion for the optical response which go beyond the TDHF is based on the following steps (Fig. 1): (i) Starting with a quantum electronic Hamiltonian [Eq. (2.1)], which represents the original quantum fermion model (QFM), we obtain the classical dynamics in the Grassman manifold M with the Poisson bracket given by Eq. (3.7) and the classical Hamiltonian defined by Eq. (3.2). This results in the classical oscillator model (COM).[36] (ii) In the vicinity of the solution $\overline{\rho}$ of the stationary HF equation (the ground state) we find the eigenmodes ξ_α of the linearized TDHF operator L which allows us to introduce a system of local coordinates z'_α [Eq. (3.13)]. We transform the coordinates z'_α [Eq. (3.21)] to make the Poisson bracket canonical to fourth-order, and obtain the classical Hamiltonian in terms of the new z variables [Eq. (3.22)]. This results in the classical canonical oscillator model (CCOM) which is equivalent to the COM, except that it uses canonical (classical boson) variables. (iii) The CCOM is quantized resulting in the quantum oscillator Hamiltonian [Eq. (D2)], which represents the quantum oscillator (boson) model (QOM). (iv) By applying the squeezed-state ansatz we obtain the Poisson bracket [Eq. (4.1)] and the classical Hamiltonian [Eqs. (E1)–(E4)] in the extended phase space of B and Y variables. The classical Liouville equation [Eq. (4.2)] then gives the desired equations of motion [Eq. (4.8)], which represent the classical dynamics of the boson coupled cluster classical model (BCCM).

It may seem at a first sight that the QOM [Eq. (D2)] constitutes an equivalent exact representation for the original fermion Hamiltonian of QFM [Eq. (2.1)]. This is however not the case. The classical oscillator Hamiltonian variables z parametrize the generalized coherent states $|\Omega\rangle$ of the $U(M)$ group in the vicinity of the HF ground state $|\Omega_0\rangle$, whereas in Eq. (3.22) z are parameters characterizing the oscillators' coherent state. The scalar product of two normalized coherent states $\langle\Omega'|\Omega\rangle$ is, therefore, different in the two cases, implying that the two systems are not equivalent on the quantum level. However since the Poisson bracket of the two is the same, the scalar products coincide to second-order in z. Therefore Eq. (D2) constitutes an approximation for the original system. The differences of the scalar product can be compensated by making a unitary transformation in a form of an exponent of a power series of the $\hat{B}$ and $\hat{B}^+$ operators which results in the bosonized quantum fermion model (BQFM) (see Fig. 1). The BQFM constitutes an equivalent representation of the QFM, however, quantum corrections to

Eq. (D2), which are responsible for the deviation of the BQFM from the QOM, can be calculated order-by-order in the oscillator operators. This procedure goes beyond the scope of this manuscript where we applied the squeezed-state ansatz to the QOM rather than to the BQFM. One consequence of this approximation is that the ground state (the initial point of ξ) is uncorrelated ($Y=0$ for $\overline{\rho}$). The present approach only includes excited state correlations. The incorporation of ground state correlations will require using the BQFM.

The resonances of the optical response within the BCCM, and therefore the transition frequencies of the system within this approximation, are determined by the harmonic Hamiltonian H_L [Eq. (4.4)] which represents a system of coupled *B*- and *Y*-oscillators. Neglecting the two-exciton *Y*-oscillators brings us back to the TDHF approximation. The interaction between the *Y*- and *B*-oscillators determined by H_{BY} [Eq. (4.7)] reduces the frequencies of the *B*-oscillators and may resolve the A_g, B_u problem.[7–10]

Finally we note that the squeezed states defined by Eq. (2.12) can be considered Gaussian wavepackets in the coordinate space. Using the oscillator coordinate representation for the Heisenberg–Weyl algebra in the space of wavefunctions $\Psi(q_1,\ldots,q_L)$ where L is the number of bosons (oscillators) the annihilation (creation) operators act as

$$\hat{B}_n=-\frac{i}{\sqrt{2}}\left(\frac{\partial}{\partial q_n}+q_n\right),\quad \hat{B}_n^+=-\frac{i}{\sqrt{2}}\left(\frac{\partial}{\partial q_n}-q_n\right). \tag{5.1}$$

Choosing the reference state Ψ_0 as the one which satisfies the conditions $\hat{B}_n\Psi_0=0$ for all $n=1,\ldots,L$ we find that Ψ_0 has a Gaussian form

$$\Psi_0(q_1\ldots,q_L)=\frac{1}{\sqrt{(2\pi)^L}}\exp\left[-\frac{1}{2}(q_1^2+\cdots+q_L^2)\right]. \tag{5.2}$$

A direct check shows that application of the operators $\hat{B}_n$, $\hat{B}_n^+$, $\hat{X}_{mn}^{(\pm)}$, and $\hat{X}_{mn}^z$ to the wavefunction preserves its Gaussian form. Action by $\hat{B}$ and $\hat{B}^+$ only shifts its center whereas the $\hat{X}$ operators change its variance as well. By acting with the elements of the extended Heisenberg–Weyl group on the reference state Ψ_0 we thus obtain a state of the form:

$$\Psi(q_1,\ldots,q_L)=A\exp\left[-\frac{1}{2}\sum_{mn}\sigma_{mn}(q_m-z_m)(q_n-z_n)\right], \tag{5.3}$$

where $z_1,\ldots,z_L$ are complex numbers which determine the average position whereas the $L\times L$ complex-valued symmetric matrix σ_{mn} (with $\sigma_{mn}=\sigma_{nm}$) determines its covariances. In particular, for $\sigma_{mn}=\delta_{mn}$ Eq. (5.3) represents an ordinary coherent state with the parameters $z_1,\ldots,z_L$. The matrix σ_{mn} is therefore responsible for squeezing. Gaussian wavepackets in Liouville space have been implemented previously in the context of the molecular dynamics in the presence of a bath.[55] Gaussian Liouville space wavepackets are squeezed Liouville space states. An extension of the present theory to include bath-induced incoherent dynamics is possible by applying the squeezed state (Gaussian) ansatz to the many-body density matrix (rather than the wavefunction) written using the oscillator representation.

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APPENDIX A: RELATIONS BETWEEN THE BILINEAR OSCILLATOR VARIABLES

In this Appendix we derive a matrix relation between the variables N_{mn} and Y_{mn} [Eq. (A7)] using the squeezed-state ansatz which allows us to eliminate the former using Eq. (2.16). We first note that it suffices to show that Eq. (A7) holds for the states Ω with $\langle\Omega|\hat{B}_i|\Omega\rangle=0$, i.e., whose wavefunctions are Gaussians centered at $q=0$. We refer to these as squeezed states. The squeezed states form an orbit M of the group G which corresponds to the algebra generated by $\hat{X}_{mn}^{(\pm)}$ and $\hat{X}_{mn}^{(z)}$. In particular this means that squeezed states are generalized coherent states with respect to G.

We shall introduce a set of functions $X_{mn}^{(\pm)}$, $X_{mn}^{(z)}$ on M

$$X_{mn}^{\pm}(\Omega)\equiv\langle\Omega|\hat{X}_{mn}^{(\pm)}|\Omega\rangle,\quad X_{mn}^{(z)}(\Omega)\equiv\langle\Omega|X_{mn}^{(z)}|\Omega\rangle, \tag{A1}$$

and define two sets of auxiliary functions

$$\begin{aligned}F_{mn}&\equiv\sum_\alpha\,[X_{ma}^{(+)}X_{an}^{(-)}-X_{ma}^{(z)}X_{an}^{(z)}],\\ G_{mn}&\equiv\sum_\alpha\,[X_{ma}^{(z)}X_{an}^{(+)}-X_{na}^{(z)}X_{am}^{(+)}].\end{aligned} \tag{A2}$$

We first prove that $G_{mn}=0$ for all m and n. To that end we note that since $[\hat{X}_{ij}^{(+)},\hat{X}_{ke}^{(+)}]=0$, the operators $\hat{X}_{ij}^{(+)}$ considered as vector fields on M determine a complex structure on M:[41] holomorphic functions f are those that satisfy the condition $\hat{X}_{ij}^{(+)}f=0$. In particular, this means that $\hat{X}_{ij}^{(+)}$ represent derivatives in the antiholomorphic directions. Since $\hat{X}_{ij}^{(+)}X_{mn}^{(+)}=0$, $X_{mn}^{(+)}$ are holomorphic functions which constitute a set of holomorphic coordinates in the vicinity of $\Omega_0\in M$ defined as the state with the wavefunction given by Eq. (5.2). A straightforward calculation shows that $X_{ij}^{(+)}(\Omega_0)=0$ and

$$X_{ij}^{(z)}(\Omega_0)=\delta_{ij}. \tag{A3}$$

A direct calculation yields $\hat{X}_{ij}^{(+)}G_{mn}=0$ which implies that G_{mn} is holomorphic and can be therefore written as a series in $X^{(+)}$ in the vicinity of the point $X_{ij}^{(+)}=0$ (i.e., Ω_0). Since $X_{ma}^z(\Omega)=\delta_{ma}$, it follows from Eq. (A2) that the expansion of G_{mn} starts with the second-order terms:

$$G_{mn}=\sum\,G_{mn;r_1s_1,r_2s_2}^{(2)}X_{r_1s_1}^{(+)}X_{r_2s_2}^{(+)}\cdots. \tag{A4}$$

We next define the degree $\deg f$ (if it exists) of a function f by $\hat{D}f=(\deg f)f$ where $\hat{D}\equiv 1/2\Sigma_j\hat{X}_{jj}^{(z)}$. In particular $\deg X_{ij}^{(\pm)}=\pm 1$, $\deg X_{ij}^{(z)}=0$, and $\deg(fg)=\deg f+\deg g$. It follows from Eq. (A2) that $\deg G_{mn}=1$. Equation (A4) on

the other hand implies that G_{mn} is a linear combination $G_{mn}=\Sigma_{j=2}^{\infty} G_{mn}^{(j)}$ with $\deg G_{mn}^{(j)}=j$ and therefore contains the degrees of 2 and higher. This implies $G_{mn}\equiv 0$ for all m and n. The proof of Eq. (2.15) can be completed by checking that $\hat{X}_{rs}^{(+)}F_{mn}$ is a linear combination of G_{ab} and hence $\hat{X}_{rs}^{(+)}F_{mn}=0$. Similarly $\hat{X}_{rs}^{(-)}F_{mn}=0$ (this can be obtained by conjugating the relation $\hat{X}_{rs}^{(+)}F_{mn}=0$). The two imply that F_{mn} is a constant. Since due to Eq. (A2) $F_{mn}(\Omega_0)=-\delta_{mn}$ we finally obtain

$$\sum_{\alpha}\left[X_{ma}^{(+)}X_{an}^{(-)}-X_{ma}^{(z)}X_{an}^{(z)}\right]=\delta_{mn}. \tag{A5}$$

It follows from Eqs. (2.9) and (2.5) that

$$Y_{mn}=X_{mn}^{(-)},\quad X_{mn}^{(z)}=N_{mn}+\tfrac{1}{2}\delta_{mn}. \tag{A6}$$

Substituting Eq. (A6) into Eq. (A5) yields the matrix equation

$$(N+\tfrac{1}{2}I)^2-Y^{+}Y=\tfrac{1}{4}I. \tag{A7}$$

Solving Eq. (A7) for N finally yields Eq. (2.16).

APPENDIX B: THE CLASSICAL OSCILLATOR MODEL

In this Appendix we map the classical limit of the fermion system onto a system of classical oscillators. We start with the Hamiltonian of Eq. (3.2) and represent ρ in Eq. (3.2) in the form of Eq. (3.10). The resulting Hamiltonian can be conveniently represented in terms of the operator t with the matrix elements t_{mn}, and the superoperator V acting in the matrix space, whose matrix elements are given in Ref. 37 in terms of $V_{mn,ke}$:

$$H(\xi)=\mathrm{Tr}\{t[\overline{\rho}+\xi+T(\xi)]\}+\mathrm{Tr}\{[\overline{\rho}+\xi+T(\xi)]\times V(\overline{\rho}+\xi+T(\xi))\}. \tag{B1}$$

Expanding $T(\xi)$ in powers of ξ

$$T(\xi)=T^{(2)}(\xi)+T^{(4)}(\xi)+\cdots, \tag{B2}$$

and keeping terms up to fourth-order and using symmetry properties of the superoperator V we obtain:

$$\begin{aligned}H(\xi)=&\mathrm{Tr}\{\overline{\rho}[t+V(\overline{\rho})]\}+\mathrm{Tr}\{\xi[t+2V(\overline{\rho})]\}\\&+\mathrm{Tr}\{T^{(2)}(\xi)[t+2V(\overline{\rho})]\}+\mathrm{Tr}[\xi V(\xi)]\\&+2\,\mathrm{Tr}[\xi V(T^{(2)}(\xi))]+\mathrm{Tr}[T^{(2)}(\xi)V(T^{(2)}(\xi))]\\&+\mathrm{Tr}\{T^{(4)}(\xi)[t+2V(\overline{\rho})]\}.\end{aligned} \tag{B3}$$

$T^{(2)}$ and $T^{(4)}$ can be calculated by expanding the exact expression

$$T(\xi)=-\left(\overline{\rho}-\frac{I}{2}\right)(I-\sqrt{I-4\xi^2}), \tag{B4}$$

obtained in Ref. 34 in powers of ξ. After some simple transformations we obtain:

$$\begin{aligned}T^{(2)}(\xi)&=-\tfrac{1}{2}[\xi,[\xi,\overline{\rho}]],\\T^{(4)}(\xi)&=-\tfrac{1}{8}[\xi,[\xi,[\xi,[\xi,\overline{\rho}]]]].\end{aligned} \tag{B5}$$

Expressing ξ in Eq. (B3) in terms of z'_{α} according to Eq. (3.14) and making use of Eq. (B5) yields:

$$\begin{aligned}H=&\frac{1}{2}\sum_{\alpha\beta}\mathrm{Tr}\{\overline{\rho}[\xi_\beta,L(\xi_\alpha)]\}z'_\alpha z'_\beta\\&+\sum_{\alpha\beta\gamma}\mathrm{Tr}\{[\overline{\rho},\xi_\alpha][V([[\xi_\beta,\overline{\rho}],\xi_\gamma]),\overline{\rho}]\}z'_\alpha z'_\beta z'_\gamma\\&+\frac{1}{4}\sum_{\alpha\beta\gamma\delta}\mathrm{Tr}\{[\overline{\rho},\xi_\alpha][V([[\xi_\gamma,\overline{\rho}],\xi_\delta]),\xi_\beta]\}z'_\alpha z'_\beta z'_\gamma z'_\delta\\&+\frac{1}{4}\sum_{\alpha\beta\gamma\delta}\mathrm{Tr}\{[\overline{\rho},\xi_\beta][V(\xi_\alpha),[[\xi_\delta,\overline{\rho}],\xi_\gamma]]\}z'_\alpha z'_\beta z'_\gamma z'_\delta\\&-\frac{1}{8}\sum_{\alpha\beta\gamma\delta}\mathrm{Tr}\{[\xi_\beta,[\xi_\gamma,[\xi_\delta,\overline{\rho}]]]L(\xi_\alpha)\}z'_\alpha z'_\beta z'_\gamma z'_\delta.\end{aligned} \tag{B6}$$

The linearlized TDHF equation operator L has the form (Ref. 37):

$$L(\xi)=[t+2V(\overline{\rho}),\xi]-2[\overline{\rho},V(\xi)]. \tag{B7}$$

In deriving Eq. (B6) we have neglected the first (constant) term in the rhs. of Eq. (B3), the second term in the rhs. of Eq. (B3) vanishes due to the stationary HF equation $[t+2V(\overline{\rho}),\overline{\rho}]=0$ which implies that $t+2V(\overline{\rho})$ contains no particle–hole components, whereas ξ belongs to the particle–hole subspace. Making use of the facts that $L(\xi_\alpha)=\Omega_\alpha\xi_\alpha$, $\Omega_{-\alpha}=-\Omega_\alpha$, the normalization conditions for the modes

$$\mathrm{Tr}(\overline{\rho}[\xi_\alpha,\xi_\beta]=\delta_{\alpha,-\beta}\,\mathrm{sgn}\,\beta, \tag{B8}$$

and an identity

$$\begin{aligned}&-\mathrm{Tr}\{[\xi_\beta,[\xi_\gamma,[\xi_\delta,\overline{\rho}]]]\xi_\alpha\}\\&\quad=\mathrm{Tr}\{[[\xi_\beta,\overline{\rho}],[\xi_\alpha,\overline{\rho}]][[\xi_\delta,\overline{\rho}],\xi_\gamma]\},\end{aligned} \tag{B9}$$

yields the classical Hamiltonian [Eq. (3.15)], with

$$\overline{V}^{(3)}_{\alpha\beta\gamma}\equiv\mathrm{Tr}\{[\overline{\rho},\xi_\alpha][V([[\xi_\beta,\overline{\rho}],\xi_\gamma]),\overline{\rho}]\}, \tag{B10}$$

$$\begin{aligned}\overline{V}^{(4)}_{\alpha\beta\gamma\delta}\equiv&\tfrac{1}{4}\mathrm{Tr}\{[\overline{\rho},\xi_\alpha][V([[\xi_\beta,\overline{\rho}],\xi_\delta]),\xi_\beta]\}\\&+\tfrac{1}{4}\mathrm{Tr}\{[\overline{\rho},\xi_\beta][V(\xi_\alpha),[[\xi_\delta,\overline{\rho}],\xi_\gamma]\}\\&+\tfrac{1}{8}\Omega_\alpha\mathrm{Tr}\{[\xi_\beta,\overline{\rho}][\xi_\alpha,\overline{\rho}][[\xi_\delta,\overline{\rho}],\xi_\gamma]\}.\end{aligned}$$

In Appendix C, we show that the canonical variables z_α are related to z'_α by means of the following transformation

$$\begin{aligned}z'_\alpha=z_\alpha-\frac{1}{8}\sum_{\beta\mu\nu}&\,\mathrm{sgn}(\alpha)\,\mathrm{Tr}\{[[\overline{\rho},\xi_{-\alpha}],[\overline{\rho},\xi_\beta]]\\&\times[[\xi_\mu,\overline{\rho}],\xi_\nu]\}z_\beta z_\mu z_\nu.\end{aligned} \tag{B11}$$

Substitution of Eq. (B11) into Eq. (3.15) and keeping the terms up to the fourth-order in z gives

$$H=\sum_{a>0}\Omega_\alpha z_\alpha^* z_\alpha+\sum_{\alpha\beta\gamma}V^{(3)}_{\alpha\beta\gamma}z_\alpha z_\beta z_\gamma + \sum_{\alpha\beta\gamma\delta}V^{(4)}_{\alpha\beta\gamma\delta}z_\alpha z_\beta z_\gamma z_\delta, \tag{B12}$$

with

$$\begin{aligned} V^{(3)}_{\alpha\beta\gamma}&=\overline{V}^{(3)}_{\alpha\beta\gamma},\\ \overline{V}^{(4)}_{\alpha\beta\gamma\delta}&=\tfrac{1}{4}\mathrm{Tr}\{[\overline{\rho},\xi_\alpha][V([[\xi_\gamma,\overline{\rho}],\xi_\delta]),\xi_\beta]\}\\ &\quad+\tfrac{1}{4}\mathrm{Tr}\{[\overline{\rho},\xi_\beta][V(\xi_\alpha),[[\xi_\delta,\overline{\rho}],\xi_\gamma]]\}\\ &\quad+\tfrac{1}{4}\Omega_\alpha\,\mathrm{Tr}\{[[[\xi_\beta,\overline{\rho}],[\xi_\alpha,\overline{\rho}]][[\xi_\delta,\overline{\rho}],\xi_\gamma]\}. \end{aligned} \tag{B13}$$

The expression for the polarization operator derived in a similar manner reads

$$P=\sum_\alpha \mu^{(1)}_\alpha z_\alpha+\sum_{\alpha\beta}\mu^{(1)}_{\alpha\beta}z_\alpha z_\beta, \tag{B14}$$

with

$$\begin{aligned} \mu^{(1)}_\alpha&=\mathrm{Tr}(\mu\xi_\alpha),.\\ \mu^{(2)}_{\alpha\beta}&=\tfrac{1}{2}\mathrm{Tr}\{\mu[[\xi_\alpha,\overline{\rho}],\xi_\beta]\}. \end{aligned} \tag{B15}$$

The Hamiltonian [Eq. (3.22)] constitutes a representation of Eq. (B12) once the summations over positive and negative Greek indices are separated. The parameters of Eq. (3.22) $V^{(n,m)}_{\alpha_{1\ldots}\alpha_n,\beta_{1\ldots}\beta_m}$ are obtained from $V^{(3)}$ and $V^{(4)}$ by performing the necessary permutations.

APPENDIX C: CANONICAL TRANSFORMATIONS AND POISSON BRACKETS

In this Appendix we first derive the Poisson bracket $\{z'_\alpha,z'_\beta\}$ to second-order, and then find a transformation to new variables z_α for which the Poisson bracket $\{z_\alpha,z_\beta\}$ is canonical to third-order [Eq. (B11)].

To calculate $\{z'_\alpha,z'_\beta\}$ we follow a procedure introduced in Ref. 37. We first note that since ρ has the form of Eq. (3.10) with

$$\xi=\sum_\alpha z'_\alpha\xi_\alpha, \tag{C1}$$

we have

$$z'_\alpha=-\mathrm{sgn}(\alpha)\,\mathrm{Tr}\{\overline{\rho}[\xi,\xi_{-\alpha}]\}. \tag{C2}$$

This implies that the function z'_α can be represented in the form

$$z'_\alpha(\rho)=\mathrm{Tr}(\rho\zeta_\alpha), \tag{C3}$$

with

$$\zeta_\alpha\equiv\mathrm{sgn}(\alpha)[\overline{\rho},\xi_{-\alpha}]. \tag{C4}$$

For such functions the Poisson bracket is given by the commutator

$$\{\zeta_\alpha,\zeta_\beta\}=i[\zeta_\alpha,\zeta_\beta]. \tag{C5}$$

Eqs. (C5) and (C3) therefore imply

$$\{z'_\alpha,z'_\beta\}(\rho)=i\,\mathrm{Tr}\{\rho[\zeta_\alpha,\zeta_\beta]\}. \tag{C6}$$

Substitution of ρ into Eq. (C6) in the form of Eq. (3.10) and taking Eq. (C1) into account yields

$$\{z'_\alpha,z'_\beta\}(\rho)=i\,\mathrm{Tr}\{\overline{\rho}[\zeta_\alpha,\zeta_\beta]\}+i\,\mathrm{Tr}\left\{[\zeta_\alpha,\zeta_\beta]T\left(\sum_\gamma z'_\gamma\xi_\gamma\right)\right\}. \tag{C7}$$

Equation (C7) constitutes an exact expression for the Poisson bracket $\{z'_\alpha,z'_\beta\}$. Substituting $T^{(2)}$ in the form of Eq. (B5) into Eq. (C7) we obtain the Poisson bracket to third-order in z'. This yields after some straightforward transformations:

$$\{z'_\alpha,z'_\beta\}=i\,\delta_{\alpha,-\beta}\,\mathrm{sgn}(\alpha)+i\sum_{\mu\nu}F_{\alpha\beta,\mu\nu}z'_\mu z'_\nu, \tag{C8}$$

with

$$\begin{aligned} F_{\alpha\beta,\mu\nu}=\frac{1}{2}\,\mathrm{sgn}(\alpha)\,\mathrm{sgn}(\beta)\,\mathrm{Tr}\{[[[\overline{\rho},\xi_{-\alpha}],[\overline{\rho},\xi_{-\beta}]]\\ \times[[\xi_\mu,\overline{\rho}],\xi_\nu]\}. \end{aligned} \tag{C9}$$

To prove Eq. (B11) we first apply the Jacobi identity for the Poisson bracket.[56] Keeping the terms up to the first order in z' we have

$$\{\{z'_\alpha,z'_\beta\},z'_\gamma\}=2\sum_\nu F_{\alpha\beta,-\gamma\nu}z'_\nu\,\mathrm{sgn}(\gamma). \tag{C10}$$

The Jacobi identity then yields

$$F_{\alpha\beta,-\gamma\nu}\,\mathrm{sgn}(\gamma)+F_{\beta\gamma,-\alpha\nu}\,\mathrm{sgn}(\alpha)+F_{\gamma\alpha,-\beta\nu}\,\mathrm{sgn}(\beta)=0. \tag{C11}$$

Taking z_α in a form

$$z_\alpha=z'_\alpha-\frac{1}{4}\sum_{\mu\nu\delta}F_{\alpha-\mu,\nu\rho}\,\mathrm{sgn}(\mu)z'_\nu z'_\nu z'_\rho, \tag{C12}$$

we make use of Eq. (C8) to obtain (retaining the terms up to second-order in z):

$$\begin{aligned} -i\{z_\alpha,z_\beta\}&=\delta_{\alpha,-\beta}\,\mathrm{sgn}(\alpha)+\sum_{\mu\nu}F_{\alpha\beta,\mu\nu}z_\mu z_\nu\\ &\quad+\frac{1}{4}\sum_{\mu\nu}F_{\alpha\beta,\mu\nu}[\mathrm{sgn}(\alpha)\,\mathrm{sgn}(-\alpha)\\ &\quad+\mathrm{sgn}(\beta)\,\mathrm{sgn}(-\beta)]z_\mu z_\nu\\ &\quad-\frac{1}{2}\sum_{\mu\nu}\mathrm{sgn}(\mu)[F_{\beta-\mu,-\alpha\nu}\,\mathrm{sgn}(\alpha)\\ &\quad+F_{-\mu\alpha,-\beta\nu}\,\mathrm{sgn}(\beta)]z_\mu x_\nu. \end{aligned} \tag{C13}$$

Applying Eq. (C11) for $\gamma=-\mu$ to the last term in the rhs of Eq. (C13) gives

$$\{z_\alpha,z_\beta\}=\delta_{\alpha,-\beta}\,\mathrm{sgn}(\alpha). \tag{C14}$$

Equation (B11) is obtained by substituting Eq. (C9) into Eq. (C12) and expressing z' in terms of z using the resulting equation, retaining terms up to third-order in z.

APPENDIX D: THE QUANTUM OSCILLATOR MODEL

In this appendix we quantize the classical oscillator Hamiltonian of Eq. (3.22) by applying the same principle used to obtain the classical limit of the fermion system. We first find a quantum oscillator Hamiltonian which yields Eq. (3.22) for its expectation values calculated on the oscillator coherent states. By introducing the oscillator annihilation (creation) operators $\hat{B}_\alpha(\hat{B}^+_\alpha)$ and reiterating that the oscillator coherent states $|z\rangle$ determined by sets of values z_γ are the eigenstates of the annihilation operators i.e., $\hat{B}_\alpha|z\rangle = z_\alpha|z\rangle$ we obtain

$$\langle z|\hat{B}^+_{\alpha_1}\cdots\hat{B}^+_{\alpha_m}\hat{B}_{\beta_1}\cdots B_{\beta_n}|z\rangle = z^*_{\alpha_1}\cdots z^*_{\alpha_m} z_{\beta_1}\cdots z_{\beta_n}. \tag{D1}$$

Equation (D1) implies that the only possible quantum Hamiltonian which satisfies the above requirement is:

$$\begin{aligned}\hat{H} = &\sum_\alpha \Omega_\alpha \hat{B}^+_\alpha \hat{B}_\alpha + \sum_{\alpha\beta\gamma} V^{(3,0)}_{\alpha\beta\gamma}\hat{B}^+_\alpha \hat{B}^+_\beta \hat{B}^+_\gamma \\ &+ \sum_{\alpha\beta\gamma} V^{(2,1)}_{\alpha\beta,\gamma}\hat{B}^+_\alpha \hat{B}^+_\beta \hat{B}_\gamma + \sum_{\alpha\beta\gamma} V^{(1,2)}_{\alpha,\beta\gamma}\hat{B}^+_\alpha \hat{B}_\beta \hat{B}_\gamma \\ &+ \sum_{\alpha\beta\gamma} V^{(0,3)}_{\alpha\beta\gamma}\hat{B}_\alpha \hat{B}_\beta \hat{B}_\gamma + \sum_{\alpha\beta\gamma\delta} V^{(4,0)}_{\alpha\beta\gamma\delta}\hat{B}^+_\alpha \hat{B}^+_\beta \hat{B}^+_\gamma \hat{B}^+_\delta \\ &+ \sum_{\alpha\beta\gamma\delta} V^{(3,1)}_{\alpha\beta\gamma,\delta}\hat{B}^+_\alpha \hat{B}^+_\beta \hat{B}_\gamma \hat{B}_\delta + \sum_{\alpha\beta\gamma\delta} V^{(2,2)}_{\alpha\beta,\gamma\delta}\hat{B}^+_\alpha \hat{B}^+_\beta \hat{B}_\gamma \hat{B}_\delta \\ &+ \sum_{\alpha\beta\gamma\delta} V^{(1,3)}_{\alpha,\beta\gamma\delta}\hat{B}^+_\alpha \hat{B}_\beta \hat{B}_\gamma \hat{B}_\delta + \sum_{\alpha\beta\gamma\delta} V^{(0,4)}_{\alpha\beta\gamma\delta}\hat{B}_\alpha \hat{B}_\beta \hat{B}_\gamma \hat{B}_\delta.\end{aligned} \tag{D2}$$

The density matrix operator $\hat{C}^+_m\hat{C}_n$ adopts the following form in terms of the oscillator operators:

$$\begin{aligned}\hat{C}^+_m\hat{C}_n = &\overline{\rho_{mn}} + \sum_\alpha [d^{(1,0)}_{mn,\alpha}\hat{B}^+_\alpha + d^{(0,1)}_{mn,\alpha}\hat{B}_\alpha] \\ &+ \frac{1}{2}\sum_{\alpha\beta}[d^{(2,0)}_{mn,\alpha\beta}\hat{B}^+_\alpha\hat{B}^+_\beta + d^{(0,2)}_{mn,\alpha\beta}\hat{B}_\alpha\hat{B}_\beta] \\ &+ \sum_{\alpha\beta} d^{(1,1)}_{mn;\alpha,\beta}\hat{B}^+_\alpha\hat{B}_\beta.\end{aligned} \tag{D3}$$

We have thus accomplished step (iii).

APPENDIX E: THE BOSON COUPLED-CLUSTER EQUATIONS

In this Appendix we carry out step (iv). By applying the squeezed-state ansatz to the resulting quantum Hamiltonian we derive closed equations of motion for the response which go beyond the TDHF. Since the wavefunction of a squeezed state is Gaussian, the Wick theorem applies and the expectation value $\langle\Psi|\hat{H}|\Psi\rangle$ can be written in terms of the B_α, $Y_{\alpha\beta}$, and $N_{\alpha\beta}$ variables introduced in Sec. II. This yields for the classical Hamiltonian in the phase space parametrized by B,Y, and N:

$$H(\tau) = H_0 + H_1 - \varepsilon(\tau)P, \tag{E1}$$

where H_0 is the classical part of the Hamiltonian, which does not contain N and Y.

$$\begin{aligned}H_0 = &\sum_\alpha \Omega_\alpha B^*_\alpha B_\alpha + \left[\sum_{\alpha\beta\gamma} V^{(3,0)}_{\alpha\beta\gamma}B^*_\alpha B^*_\beta B^*_\gamma + \sum_{\alpha\beta\gamma} V^{(2,1)}_{\alpha\beta,\gamma}B^*_\alpha B^*_\beta B_\gamma + \text{c.c.}\right] + \left[\sum_{\alpha\beta\gamma\delta} V^{(4,0)}_{\alpha\beta\gamma\delta}B^*_\alpha B^*_\beta B^*_\gamma B^*_\delta\right. \\ &\left. + \sum_{\alpha\beta\gamma\delta} V^{(3,1)}_{\alpha\beta\gamma,\delta}B^*_\alpha B^*_\beta B^*_\gamma B_\delta + \text{c.c.}\right] + \sum_{\alpha\beta\gamma\delta} V^{(2,2)}_{\alpha\beta,\gamma\delta}B^*_\alpha B^*_\beta B_\gamma B_\delta.\end{aligned} \tag{E2}$$

H_1 contains the variables Y and N and, therefore, represents the quantum corrections:

$$\begin{aligned}H_1 = &\sum_\alpha \Omega_\alpha N_{\alpha\alpha} + \left[3\sum_{\alpha\beta\gamma} V^{(3,0)}_{\alpha\beta\gamma}B^*_\alpha Y_{\beta\gamma} + \text{c.c.}\right] + \left[\sum_{\alpha\beta\gamma} V^{(2,1)}_{\alpha\beta,\gamma}Y^*_{\alpha\beta}B_\gamma + 2\sum_{\alpha\beta\gamma} V^{(2,1)}_{\alpha\beta,\gamma}B^*_\alpha N_{\beta\gamma} + \text{c.c.}\right] \\ &+ \left[6\sum_{\alpha\beta\gamma\delta} V^{(4,0)}_{\alpha\beta\gamma\delta}B^*_\alpha B^*_\beta Y^*_{\gamma\delta} + 6\sum_{\alpha\beta\gamma\delta} V^{(4,0)}_{\alpha\beta\gamma\delta}Y^*_{\alpha\beta}Y^*_{\gamma\delta} + \text{c.c.}\right] + \left[3\sum_{\alpha\beta\gamma\delta} V^{(3,1)}_{\alpha\beta\gamma,\delta}B^*_\alpha B_\delta Y^*_{\beta\gamma} + 3\sum_{\alpha\beta\gamma\delta} V^{(3,1)}_{\alpha\beta\gamma,\delta}B^*_\alpha B^*_\beta N_{\gamma\delta}\right. \\ &\left. + 3\sum_{\alpha\beta\gamma\delta} V^{(3,1)}_{\alpha\beta\gamma,\delta}Y^*_{\alpha\beta}N_{\gamma\delta} + \text{c.c.}\right] + \left[\sum_{\alpha\beta\gamma\delta} V^{(2,2)}_{\alpha\beta,\gamma\delta}B^*_\alpha B^*_\beta Y_{\gamma\delta} + \text{c.c.}\right] \\ &+ 4\sum_{\alpha\beta\gamma\delta} V^{(2,2)}_{\alpha\beta,\gamma\delta}B^*_\alpha B_\gamma N_{\beta\delta} + \sum_{\alpha\beta\gamma\delta} V^{(2,2)}_{\alpha\beta,\gamma\delta}Y^*_{\alpha\beta}Y_{\gamma\delta} + 4\sum_{\alpha\beta\gamma\delta} V^{(2,2)}_{\alpha\beta,\gamma\delta}N_{\alpha\gamma}N_{\beta\delta}.\end{aligned} \tag{E3}$$

The polarization similarly adopts the form

$$P = \left[\sum_\alpha \mu^{(1,0)}_\alpha B^*_\alpha + \text{c.c.}\right] + \frac{1}{2}\left[\sum_{\alpha\beta} \mu^{(2,0)}_{\alpha\beta}B^*_\alpha B^*_\beta + \text{c.c.}\right] + \frac{1}{2}\left[\sum_{\alpha\beta} \mu^{(2,0)}_{\alpha\beta}Y^*_{\alpha\beta} + \text{c.c.}\right] + \sum_{\alpha\beta} \mu^{(1,1)}_{\alpha,\beta}B^*_\alpha B_\beta + \sum_{\alpha\beta} \mu^{(1,1)}_{\alpha,\beta}N_{\alpha\beta}. \tag{E4}$$

The equations of motion are obtained by substituting the Hamiltonian given by Eqs. (D4)–(D7) and the Poisson bracket [Eq. (4.1)] into the classical Liouville equation [Eq. (4.2)]. This yields the equations of motion in the form of Eq. (4.8) with the nonlinear terms $\dot{B}$ and $\dot{Y}$ in the form:

$$\dot{B}_\alpha = \dot{B}_\alpha^{(2)} + \dot{B}_\alpha^{(3)} + \dot{B}_\alpha^{(\varepsilon)}$$
$$\dot{Y}_{\mu\nu} = \dot{Y}_{\mu\nu}^{(2)} + \dot{Y}_{\mu\nu}^{(3)} + \dot{Y}_{\mu\nu}^{(\varepsilon)}, \tag{E5}$$

where $\dot{B}^{(j)}$ and $\dot{Y}^{(j)}$ denote the jth order terms in the corresponding equations whereas $\dot{B}^{(\varepsilon)}$ and $\dot{Y}^{(\varepsilon)}$ stand for the terms that contain the deriving field. The B terms are given by

$$\dot{B}_\alpha^{(2)} = 3\sum_{\beta\gamma} V_{\alpha\beta\gamma}^{(3,0)} B_\beta^* B_\gamma^* + 2\sum_{\beta\gamma} V_{\alpha\beta,\gamma}^{(2,1)} B_\beta^* B_\gamma + \sum_{\beta\gamma} V_{\alpha,\beta\gamma}^{(1,2)} B_\beta B_\gamma + 2\sum_{\beta\gamma\delta} V_{\alpha\beta,\gamma}^{(2,1)} Y_{\beta\delta}^* Y_{\gamma\delta} + 12\sum_{\beta\gamma\delta} V_{\alpha\beta\gamma\delta}^{(4,0)} B_\beta^* Y_{\gamma\delta}^* + 3\sum_{\delta\gamma\delta} V_{\alpha\beta\gamma,\delta}^{(3,1)} B_\delta Y_{\beta\gamma}^* + 2\sum_{\beta\gamma\delta} V_{\alpha\beta,\gamma\delta}^{(2,2)} B_\beta^* Y_{\gamma\delta} + 3\sum_{\beta\gamma\delta} V_{\alpha,\beta\gamma\delta}^{(1,3)} B_\beta Y_{\gamma\delta},$$

$$\dot{B}_\alpha^{(3)} = 4\sum_{\beta\gamma\delta} V_{\alpha\beta\gamma\delta}^{(4,0)} B_\beta^* B_\gamma^* B_\delta^* + 3\sum_{\beta\gamma\delta} V_{\alpha\beta\gamma,\delta}^{(3,1)} B_\beta^* B_\gamma^* B_\delta + 2\sum_{\beta\gamma\delta} V_{\alpha\beta,\gamma\delta}^{(2,2)} B_\beta^* B_\gamma B_\delta + \sum_{\beta\gamma\delta} V_{\alpha,\beta\gamma\delta}^{(1,3)} B_\beta B_\gamma B_\delta + 6\sum_{\beta\gamma\delta\mu} V_{\alpha\beta\gamma,\delta}^{(3,1)} B_\beta^* Y_{\mu\gamma}^* Y_{\mu\delta} + 4\sum_{\beta\gamma\delta\mu} V_{\alpha\beta,\gamma\delta}^{(2,2)} B_\gamma Y_{\mu\beta}^* Y_{\mu\delta},$$

$$\dot{B}_\alpha^{(\varepsilon)} = -\mu_\alpha^{(1,0)}\varepsilon - \sum_\beta \mu_{\alpha\beta}^{(2,0)} B_\beta^* \varepsilon - \sum_\beta \mu_{\alpha,\beta}^{(1,1)} B_\beta \varepsilon.$$

The corresponding terms for Y are

$$\dot{Y}_{\mu\nu}^{(2)} = 6\sum_{\gamma\delta\alpha} V_{\mu\nu\gamma,\delta}^{(3,1)} Y_{\gamma\alpha}^* Y_{\delta\alpha} + 3\sum_{\alpha\beta\delta} V_{\alpha\beta\mu,\delta}^{(3,1)} Y_{\alpha\beta}^* Y_{\delta\nu} + 3\sum_{\alpha\beta\delta} V_{\alpha\beta\nu,\delta}^{(3,1)} Y_{\alpha\beta}^* Y_{\delta\mu} + 2\sum_{\gamma\delta} V_{\mu\nu,\gamma\delta}^{(2,2)} B_\gamma B_\delta + 6\sum_{\alpha\delta} V_{\alpha\mu\nu,\delta}^{(3,1)} B_\alpha^* B_\delta + 12\sum_{\alpha\beta} V_{\mu\nu\alpha\beta}^{(4,0)} B_\alpha^* B_\beta^* + 2\sum_{\alpha\gamma} V_{\alpha\mu,\gamma}^{(2,1)} B_\alpha^* Y_{\nu\gamma} + 2\sum_{\alpha\gamma} V_{\alpha\nu,\gamma}^{(2,1)} B_\alpha^* Y_{\mu\gamma} + 2\sum_{\beta\gamma} V_{\mu,\beta\gamma}^{(1,2)} B_\gamma Y_{\nu\beta} + 2\sum_{\beta\gamma} V_{\nu,\beta\gamma}^{(1,2)} B_\gamma Y_{\mu\beta},$$

$$\dot{Y}_{\mu\nu}^{(3)} = 2\sum_{\beta\gamma\delta\alpha} V_{\mu\beta,\gamma\delta}^{(2,2)} Y_{\beta\alpha}^* Y_{\nu\alpha} Y_{\gamma\delta} + 2\sum_{\beta\gamma\delta\alpha} V_{\nu\beta,\gamma\delta}^{(2,2)} Y_{\beta\alpha}^* Y_{\mu\alpha} Y_{\gamma\delta} + 8\sum_{\beta\gamma\delta\alpha} V_{\mu\beta,\gamma\delta}^{(2,2)} Y_{\beta\alpha}^* Y_{\delta\alpha} Y_{\nu\gamma} + 8\sum_{\beta\gamma\delta\alpha} V_{\nu\beta,\gamma\delta}^{(2,2)} Y_{\beta\alpha}^* Y_{\delta\alpha} Y_{\mu\gamma} + 12\sum_{\alpha\beta\delta\gamma} V_{\alpha\beta\mu\delta}^{(4,0)} Y_{\alpha\beta}^* Y_{\delta\gamma}^* Y_{\nu\gamma} + 12\sum_{\alpha\beta\delta\gamma} V_{\alpha\beta\nu\delta}^{(4,0)} Y_{\alpha\beta}^* Y_{\delta\gamma}^* Y_{\delta\mu} + 6\sum_{\alpha\gamma\delta} V_{\alpha\mu\gamma}^{(3,0)} B_\alpha^* Y_{\gamma\delta}^* Y_{\nu\delta} + 6\sum_{\alpha\gamma\delta} V_{\alpha\nu\gamma}^{(3,0)} B_\alpha^* Y_{\gamma\delta}^* Y_{\mu\delta} + 2\sum_{\beta\gamma\delta} V_{\mu\beta,\gamma}^{(2,1)} Y_{\beta\delta}^* Y_{\nu\delta} B_\gamma + 2\sum_{\beta\gamma\delta} V_{\nu\beta,\gamma}^{(2,1)} Y_{\beta\delta}^* Y_{\mu\delta} B_\gamma + 3\sum_{\alpha\beta\delta} V_{\alpha\beta\mu,\delta}^{(3,1)} B_\alpha^* B_\beta^* Y_{\nu\delta} + 3\sum_{\alpha\beta\delta} V_{\alpha\beta\nu,\delta}^{(3,1)} B_\alpha^* B_\beta^* Y_{\mu\delta} + 4\sum_{\alpha\gamma\delta} V_{\alpha\mu,\gamma\delta}^{(2,2)} B_\alpha^* B_\gamma Y_{\nu\delta} + 4\sum_{\alpha\gamma\delta} V_{\alpha\nu,\gamma\delta}^{(2,2)} B_\alpha^* B_\gamma Y_{\mu\delta} + \sum_{\alpha\gamma\delta} V_{\mu,\beta\gamma\delta}^{(1,3)} B_\gamma B_\delta Y_{\nu\beta} + \sum_{\beta\gamma\delta} V_{\nu,\beta\gamma\delta}^{(1,3)} B_\gamma B_\delta Y_{\mu\beta}. \tag{E7}$$

Finally, the terms involving the driving field are:

$$\dot{Y}_{\mu\nu}^{(\varepsilon)} = -\mu_{\mu\nu}^{(2,0)}\varepsilon - \sum_\beta [\mu_{\mu,\beta}^{(1,1)} Y_{\beta\nu} + \mu_{\nu,\beta}^{(1,1)} Y_{\beta\mu}]\varepsilon - \sum_{\alpha\beta} [\mu_{\alpha\mu}^{(2,0)} Y_{\alpha\beta}^* Y_{\nu\beta} + \mu_{\alpha\nu,\beta}^{(2,0)} Y_{\alpha\beta}^* Y_{\mu\beta}]\varepsilon. \tag{E8}$$

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