Generalized coherent-state derivation of time-dependent density-functional theory equations for superconductors

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Equations of motion are derived for the normal and anomalous single-electron density matrices of a Fermi liquid using a time-dependent finite-temperature generalized coherent-state variational ansatz for the many-body density matrix. Self-consistent equations for the order parameter Δ allow us to investigate the interplay of the Coulomb repulsion and pairing attraction in homogeneous and inhomogeneous Fermi liquids with spontaneously broken symmetry such as high-temperature superconductors. The temperature of the Kosterlitz-Thouless transition to the two-dimensional superfluidity is calculated.

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I. INTRODUCTION

The BCS ansatz for the ground-state wave function of superconductors predicts a gap in the spectrum[1] originating from the pairing of two electrons with opposite momenta and spin projections. This pairing is induced by the effective attraction induced by electron-phonon interaction. The standard BCS description of superconductivity, as well as Eliashberg's extension to incorporate Coulomb repulsion between electrons[2,3] does not apply for strongly correlated electrons such as in high-temperature superconductors [$YBa_2Cu_3O_{7-\delta}$ (YBCO)].[4] In two-dimensional (2D) superconductors (e.g., in cuprates),[4,5] the phase transition to the superfluid state takes place at temperatures below the mean-field phase transition temperature for the appearance of the gap,[6,7] similar to the Kosterlitz-Thouless phase transition. The important role played by electron-electron correlations in the local order parameter above the phase transition temperature, while the system is still in the normal phase, was discussed.[5]

Superconductivity in strongly correlated electron systems is often described using effective Hubbard Hamiltonians.[4] Taking electron-electron exchange and correlations into account, the ground-state energies and collective excitation spectrum of superconductors have been calculated by density-functional theory (DFT) or its time-dependent extension (TDDFT).[8–11] In the Oliveira-Gross-Kohn (OGK) DFT equations[8–11] the normal $\rho(\mathbf{r},\mathbf{r}')$ and anomalous $\sigma(\mathbf{r},\mathbf{r}')$ density matrices satisfy the generalized Bogoliubov–de Gennes equations[8,12] which include one external field $v_s(\mathbf{r})$ coupled to the normal charge density $n(\mathbf{r})=\rho(\mathbf{r},\mathbf{r})$ and a second field $\Delta(\mathbf{r},\mathbf{r}')$ coupled to $\sigma(\mathbf{r},\mathbf{r}')$. These fields contain exchange-correlation potentials $v_{xc}(\mathbf{r})$ and $\Delta_{xc}(\mathbf{r},\mathbf{r}')$, respectively, obtained by requiring the charge density $\rho(\mathbf{r})$ and anomalous density matrix $\sigma(\mathbf{r},\mathbf{r}')$ of the interacting and noninteracting systems to be identical. TDDFT requires the same conditions to be satisfied for an externally driven system at all times.[9] The zero-order approximation in the standard DFT obtained by neglecting the exchange-correlation fields results in the Bogoliubov–de Gennes equations[8,12] which take into account the BCS pairing and classical Coulomb correlations at the Hartree level.

The DFT equations for superconductors are usually formulated for the Kohn-Sham orbitals in Hilbert space and constitute a system of four self-consistent equations for Bogoliubov transformation coefficients $u(\mathbf{r})$ and $v(\mathbf{r})$, the normal density $n(\mathbf{r})$, and the anomalous density matrix $\sigma(\mathbf{r},\mathbf{r}')$.[8] An extension of these equations to include a magnetic field was proposed as well.[11]

In this article we present an alternative derivation of the OGK equations for a Fermi liquid with spontaneously broken symmetry (FLSBS) based on a generalized coherent-state (GCS) ansatz for the many-electron density matrix. Coherent states were first used to describe anharmonic dynamical systems such as many-body interacting fermions and bosons[13] while preserving some of the useful properties of the original Glauber's coherent states for the harmonic oscillator.[14,15] They encompass the Glauber coherent states as well as the squeezed states.[14,15] The time-dependent Hartree-Fock-Bogoliubov (TDHFB) equations were derived for boson systems using the GCS ansatz.[16,17] GCS's are particularly suitable for variational dynamics by virtue the underlying Lie group algebra.[13,18] Using this ansatz, we derive equations of motion for expectation values of the normal and anomalous density matrices. We use the time-dependent variational principle which allows description of the many-body system in terms of a small number of parameters and is formally closely related to classical Poisson bracket mechanics—i.e., to the variational equations of motion derived from the minimum-action principle. A GCS representation for the BCS wave function has been used to analyze the coexistence of superconductivity and ferroelectricity.[19,20] We obtain self-consistent equations of motion for the normal and anomalous density matrices[21] and derive the spectrum of collective excitations, the density of the superfluid component at finite temperatures, and the temperature of the transition to the superfluid state for homogeneous and inhomogeneous superconductors.

Reduced descriptions of many-body systems are naturally recast using density matrices,[22,23] and we found it useful to adopt the Liouville space density matrix representation[22,24] of TDDFT (Ref. 8,9, and 25) for the normal and anomalous density matrices. This requires solving only two equations of motion for the normal and anomalous density matrices

$\rho(\mathbf{r},\mathbf{r}')$ and $\sigma(\mathbf{r},\mathbf{r}')$ coupled to two artificial external fields. These fields contain an exchange-correlation contribution and guarantee the charge density and anomalous density matrix to be exact at all times.[8,9] The ground state is the stationary solution of these equations of motion. This is in contrast to the four self-consistent equations for Bogoliubov coefficients $u(\mathbf{r})$ and $v(\mathbf{r})$ and the charge density $n(\mathbf{r})=\rho(\mathbf{r},\mathbf{r})$ and anomalous density matrix $\sigma(\mathbf{r},\mathbf{r}')$.[12,26] The density matrix is a two-point function compared to two one-point functions u and v (Refs. 8 and 9); nevertheless, the computational cost can be reduced. The reason is that the density matrices have nonvanishing elements only when $|\mathbf{r}-\mathbf{r}'|$ is less than a coherence size, which is typically very short. This allows us to neglect many density matrix elements, making its size scaling linear rather then quadratic. The Liouville space representation provides a clear picture of the underlying coherence, since it is not possible to include the coherence size explicitly in the traditional Hilbert space computations.

Using our GCS ansatz we further define expressions for first-order adiabatic (time-independent) exchange-correlation contributions to the order parameter, which include electron exchange in the spectrum. This provides corrections to the charge density and the anomalous density matrix of Ref. 26 at each order in the perturbative series for the exchange-correlation potential. The present approach is applicable for superconductors in general, but it is particularly relevant to strongly correlated high-T_c superconductors, where the BCS and Eliashberg theories do not apply.

II. COHERENT-STATE FREE ENERGY

We start with the many-electron Hamiltonian $\hat{H}$ where the electron-electron interaction consists of both Coulomb repulsion $V(\mathbf{r}-\mathbf{r}')=e^2/|\mathbf{r}-\mathbf{r}'|$ and pairing attraction $W(\mathbf{r}-\mathbf{r}')$ between two electrons with opposite spins[8]:

$$\begin{aligned}\hat{H}=&\sum_{\nu}\int d\mathbf{r}\,\hat{\psi}_{\nu}^{\dagger}(\mathbf{r})\left(-\frac{1}{2m}\nabla_{\mathbf{r}}^2-\mu\right)\hat{\psi}_{\nu}(\mathbf{r})\\&+\frac{1}{2}\sum_{\nu\nu'}\int d\mathbf{r}\int d\mathbf{r}'\,\hat{\psi}_{\nu}^{\dagger}(\mathbf{r})\hat{\psi}_{\nu'}^{\dagger}(\mathbf{r}')\\&\times V(\mathbf{r}-\mathbf{r}')\hat{\psi}_{\nu'}(\mathbf{r}')\hat{\psi}_{\nu}(\mathbf{r})\\&-\sum_{\nu,\nu'\neq\nu}\int d\mathbf{r}\int d\mathbf{r}'\,\hat{\psi}_{\nu}^{\dagger}(\mathbf{r})\hat{\psi}_{\nu'}^{\dagger}(\mathbf{r}')\\&\times W(\mathbf{r}-\mathbf{r}')\hat{\psi}_{\nu'}(\mathbf{r}')\hat{\psi}_{\nu}(\mathbf{r}).\end{aligned}\tag{1}$$

Here $\hat{\psi}^{\dagger}(\mathbf{r})$ and $\hat{\psi}(\mathbf{r})$ are the Fermi creation and annihilation field operators with the anticommutation relations $[\hat{\psi}(\mathbf{r}),\hat{\psi}^{\dagger}(\mathbf{r}')]_+=\delta(\mathbf{r}-\mathbf{r}')$ and $[\hat{\psi}(\mathbf{r}),\hat{\psi}(\mathbf{r}')]_+=[\hat{\psi}^{\dagger}(\mathbf{r}),\hat{\psi}^{\dagger}(\mathbf{r}')]_+=0$, and the indices ν and ν' denote the spin projections; m is the effective band mass of electron, μ is the chemical potential (Fermi energy), and e is the electron charge.

We further expand the field operators in a single-electron basis set $\phi_{i\nu}(\mathbf{r})$:

$$\hat{\psi}_{\nu}^{\dagger}(\mathbf{r},t)=\sum_i\,\phi_{i\nu}^{*}(\mathbf{r})\hat{a}_{i\nu}^{\dagger}(t),$$

$$\hat{\psi}_{\nu}(\mathbf{r},t)=\sum_i\,\phi_{i\nu}(\mathbf{r})\hat{a}_{i\nu}(t),\tag{2}$$

where $\hat{a}_{i\nu}^{\dagger}$ and $\hat{a}_{i\nu}$ are the corresponding Fermi operators with the anticommutation relations $[\hat{a}_i,\hat{a}_j]_+=\delta_{ij}$ and $[\hat{a}_i,\hat{a}_j]_+=[\hat{a}_i^{\dagger},\hat{a}_j^{\dagger}]_+=0$, $\phi_{i\nu}(\mathbf{r})$ are orthonormal atomic basis functions, and i runs over all basis electronic orbitals.

Substituting Eq. (2) into Eq. (1) gives

$$\begin{aligned}\hat{H}=&\sum_{i,j,\nu}t_{ij}\hat{a}_{i\nu}^{\dagger}\hat{a}_{j\nu}+\sum_{\substack{i,j,k,l\\\nu,\nu'}}V_{ijkl}\hat{a}_{i\nu}^{\dagger}\hat{a}_{j\nu'}^{\dagger}\hat{a}_{k\nu'}\hat{a}_{l\nu}\\&-\sum_{\substack{i,j,k,l\\\nu,\nu'\neq\nu}}W_{ijkl}\hat{a}_{i\nu}^{\dagger}\hat{a}_{j\nu'}^{\dagger}\hat{a}_{k\nu'}\hat{a}_{l\nu}.\end{aligned}\tag{3}$$

Here t_{ij} is the single-electron matrix element,

$$t_{ij}=\int d\mathbf{r}\phi_i^{*}(\mathbf{r})\left(-\frac{\hbar^2\nabla_{\mathbf{r}}^2}{2m_b}-\mu\right)\phi_j(\mathbf{r}),\tag{4}$$

V_{ijkl} is the Coulomb electron-electron repulsion,

$$V_{ijkl}=\int d\mathbf{r}_1 d\mathbf{r}_2\phi_i^{*}(\mathbf{r}_1)\phi_j^{*}(\mathbf{r}_2)\frac{e^2}{|\mathbf{r}_1-\mathbf{r}_2|}\phi_k(\mathbf{r}_1)\phi_l(\mathbf{r}_2),\tag{5}$$

and W_{ijkl} is an attraction responsible for the creation of electron Cooper pair.

Equation (3) describes the interacting many-fermion system with Coulomb repulsion and attraction between two electrons with opposite projections of spin. In ordinary (BCS) superconductors this attraction originates from an electron-phonon interaction[1]; in YBCO superconductors the short-range attraction results from the thermodynamically equilibrated phase ordering producing charge stripe order,[4] and it assumes in the following form:

$$\begin{aligned}W_{ijkl}=&A\int d\mathbf{r}_1 d\mathbf{r}_2\phi_i^{*}(\mathbf{r}_1)\phi_j^{*}(\mathbf{r}_2)\\&\times\left(\frac{|\mathbf{r}_1-\mathbf{r}_2|}{r_0}\right)^{-n}\phi_k(\mathbf{r}_1)\phi_l(\mathbf{r}_2),\end{aligned}\tag{6}$$

where $n>1$ is a positive rational number; A and r_0 are constants, determined by system geometry.

Our derivation is based on the following ansatz for the time-dependent many-electron density matrix. At zero temperature the system is in a pure state, and the density matrix is given by $K(t)\propto|\psi(t)\rangle\langle\psi(t)|$, where the (unnormalized) many-electron wave function is assumed to be of the form

$$|\psi(t)\rangle = \exp\Bigg[\int dt \int d\mathbf{r} \int d\mathbf{r}' \sum_{\nu\nu'} h(\mathbf{r},\mathbf{r}',t)\hat{\psi}_\nu(\mathbf{r})\hat{\psi}^\dagger_{\nu'}(\mathbf{r}') + \sum_{\nu,\nu'\neq\nu} \Delta(\mathbf{r},\mathbf{r}',t)\hat{\psi}^\dagger_\nu(\mathbf{r})\hat{\psi}^\dagger_{\nu'}(\mathbf{r}') \Bigg] |\Omega_0\rangle$$

$$= \exp\Bigg[\sum_{ij} \Bigg(\sum_{\nu\nu'} h_{ij}(t)\hat{a}^\dagger_{i\nu}\hat{a}_{j\nu'} + \sum_{\nu,\nu'\neq\nu} \Delta_{ij}(t)\hat{a}^\dagger_{i\nu}\hat{a}^\dagger_{j\nu'} \Bigg) \Bigg] |\Omega_0\rangle, \tag{7}$$

with $|\Omega_0\rangle$ being an arbitrary single Slater determinant.[27] Equation (7) is a generalization of the random phase approximation (RPA) and BCS wave functions: setting $\Delta_{ij}=0$ it reduces to the Thouless representation of the single Slater determinant[27], for $h_{ij}=0$ it reduces to the BCS ansatz for the superconductor.[1]

At finite temperature T our ansatz reads

$$K = \frac{\exp[-\hat{H}_0/(k_B T)]}{\mathrm{Tr}\,\exp[-\hat{H}_0/(k_B T)]}, \tag{8}$$

where k_B is a Boltzmann constant and

$$\hat{H}_0 = \int dt \int d\mathbf{r} \int d\mathbf{r}' \Bigg[\sum_{\nu\nu'} h(\mathbf{r},\mathbf{r}',t)\hat{\psi}_\nu(\mathbf{r})\hat{\psi}^\dagger_{\nu'}(\mathbf{r}') + h^*(\mathbf{r},\mathbf{r}',t)\hat{\psi}^\dagger_\nu(\mathbf{r})\hat{\psi}_{\nu'}(\mathbf{r}') + \sum_{\nu,\nu'\neq\nu} \Delta(\mathbf{r},\mathbf{r}',t)\hat{\psi}^\dagger_\nu(\mathbf{r})\hat{\psi}^\dagger_{\nu'}(\mathbf{r}') + \sum_{\nu,\nu'\neq\nu} \Delta^*(\mathbf{r},\mathbf{r}',t)\hat{\psi}_\nu(\mathbf{r})\hat{\psi}_{\nu'}(\mathbf{r}') \Bigg] \tag{9}$$

or using our basis set

$$\hat{H}_0 = \sum_{ij} \Bigg[\sum_{\nu\nu'} h_{ij}(t)\hat{a}^\dagger_{i\nu}\hat{a}_{j\nu'} + \sum_{\nu\nu'} h^*_{ij}(t)\hat{a}_{i\nu}\hat{a}^\dagger_{j\nu'} + \sum_{\nu,\nu'\neq\nu} \Delta_{ij}(t)\hat{a}^\dagger_{i\nu}\hat{a}^\dagger_{j\nu'} + \sum_{\nu,\nu'\neq\nu} \Delta^*_{ij}(t)\hat{a}_{i\nu}\hat{a}_{j\nu'} \Bigg]. \tag{10}$$

Here "Tr" denotes the trace in the many-electron Fock space. A similar ansatz was recently used for Bose condensation.[17] Equation (8) constitutes a GCS (Ref. 28 and 29) [see Eq. (A1)]. Equation (7) is a limiting case of Eq. (8) obtained by a specific choice of parameters[17]; Eq. (8) thus holds at finite temperatures as well as at $T=0$.

The parameters h_{ij} and Δ_{ij} will be determined variationally by minimizing the grand canonical free energy

$$F(\mu,T) \equiv \mathrm{Tr}(\hat{H}K) - k_B T \mathrm{Tr}[K\ln(K)] \equiv \mathcal{H} - T\mathcal{S}. \tag{11}$$

Here $\mathcal{H}$ is the enthalpy, $\mathcal{S}$ is the entropy, and the chemical potential μ controls the average number of electrons N through the following constraint[17,30]:

$$\mathrm{Tr}\Bigg(K \sum_{i\nu} \hat{a}^\dagger_{i\nu}\hat{a}_{i\nu} \Bigg) = N. \tag{12}$$

Instead of using h and Δ as the variational parameters, we shall switch to the following variables: the normal density matrix

$$\rho_{i\nu j\nu} \equiv \mathrm{Tr}(K\hat{a}^\dagger_{i\nu}\hat{a}_{j\nu}) \tag{13}$$

and the anomalous density matrices

$$\sigma_{i\nu j-\nu} \equiv \mathrm{Tr}(K\hat{a}_{i\nu}\hat{a}_{j-\nu}), \tag{14}$$

$$\sigma^*_{i\nu j-\nu} \equiv Tr(K\hat{a}^\dagger_{i\nu}\hat{a}^\dagger_{j-\nu}). \tag{15}$$

The next step will be to express the free energy in terms of ρ and σ. We start with the enthalpy. Since K is the exponent of a quadratic operator, we can use Wick's theorem[21] and express all averages of products of creation and annihilation operators with respect to K (denoted with a subscript 0) as averages of pairs of operators, which are generators of the closed algebra. In particular we have

$$\langle \hat{a}^\dagger_i\hat{a}^\dagger_j\hat{a}_k\hat{a}_m\rangle_0 = -\langle \hat{a}^\dagger_i\hat{a}_k\rangle_0\langle \hat{a}^\dagger_j\hat{a}_m\rangle_0 + \langle \hat{a}^\dagger_j\hat{a}_k\rangle_0\langle \hat{a}^\dagger_i\hat{a}_m\rangle_0 + \langle \hat{a}^\dagger_i\hat{a}^\dagger_j\rangle_0\langle \hat{a}_k\hat{a}_m\rangle_0. \tag{16}$$

Using this factorization we obtain for the enthalpy (see the Appendix), where for brevity we omit spin indices ν:

$$\mathcal{H} \equiv \mathrm{Tr}(\hat{H}K) = \frac{1}{2}\sum_{i,j} [\tilde{h}_{ij}(\rho_{ij} - \rho^*_{ji}) + \tilde{\Delta}_{ij}\sigma^*_{ij} + \tilde{\Delta}^*_{ij}\sigma_{ji}]. \tag{17}$$

Here $\tilde{h}$ is a matrix with elements

$$\tilde{h}_{ij} = t_{ij} + \frac{1}{2U}\sum_{\substack{k,l \\ \nu,\nu'}} (V_{iklj} - \delta_{\nu\nu'}V_{ilkj})\rho_{k\nu l\nu}. \tag{18}$$

U is the volume and $\tilde{\Delta}$ is the order parameter matrix with elements

$$\tilde{\Delta}_{ij} \equiv \sum_{mn} W_{ijmn}\sigma_{mn}. \tag{19}$$

We next require the expectation of the effective Hamiltonian $\hat{H}_0$ [Eq. (10)] to be the same as the expectation of $\hat{H}$ [Eq. (3)]:

$$\mathrm{Tr}(K\hat{H}) = \mathrm{Tr}(K\hat{H}_0). \tag{20}$$

It can be easily verified that the condition, Eq. (20), is met provided we set in Eq. (10) $h_{ij}=\tilde{h}_{ij}$ [Eq. (18)] and $\Delta_{ij}=\tilde{\Delta}_{ij}$ [Eq. (19)].[28] We next turn to computing the enthalpy. The effective quadratic Hamiltonian $\hat{H}_0$ given by Eq. (10) can be alternatively recast in the form

$$\hat{H}=\mathrm{tr}(\hat{Q}\hat{R}), \tag{21}$$

where the matrix $\hat{Q}$ is defined as

$$\hat{Q}\equiv\begin{pmatrix} \hat{h} & \hat{\Delta} \\ -\hat{\Delta}^* & -\hat{h}^* \end{pmatrix}. \tag{22}$$

$\hat{R}$ is the generalized single-particle density matrix,

$$\hat{R}\equiv\begin{pmatrix} \hat{\rho} & \hat{\sigma} \\ -\hat{\sigma}^* & -\hat{\rho}^*+\mathbb{1} \end{pmatrix}, \tag{23}$$

and the symbol "tr" in Eq. (21) stands for the trace in the single-electron space.

Using the ansatz, Eq. (8), the many-electron system described by the Hamiltonian, Eq. (3), can be mapped onto the ideal system of noninteracting fermions with the quadratic Hamiltonian $\hat{H}_0$, determined by the matrix $\hat{Q}$,[28] with the parameters defined by Eqs. (18) and (19). The entropy $\mathcal{S}$ of the system of fermions described by the quadratic Hamiltonian, Eq. (21), is given by[28]

$$\mathcal{S}(\hat{\rho},\hat{\sigma},\mu,T)=-k_B\mathrm{tr}[\hat{f}\ln\hat{f}+(\mathbb{1}-\hat{f})\ln(\mathbb{1}-\hat{f})], \tag{24}$$

where the matrix $\hat{f}$ is

$$\hat{f}\equiv\frac{1}{\exp[\hat{Q}/(k_BT)]+\mathbb{1}}. \tag{25}$$

We are looking for the normal and anomalous density matrices $\hat{\rho}$ and $\hat{\sigma}$ that minimize the free energy [Eq. (11)] together with Eqs. (17) and (24) assuming the GCS ansatz for the many-electron density matrix, Eq. (8). This minimization yields the following equation[28]:

$$\hat{R}=\frac{1}{\exp[\hat{Q}/(k_BT)]+\mathbb{1}}, \tag{26}$$

which gives

$$\hat{\rho}=1-\hat{\varepsilon}[\hat{E}]^{-1}\tanh[\hat{E}/(2k_BT)], \tag{27}$$

$$\hat{\sigma}=\frac{1}{2}\hat{\Delta}[\hat{E}]^{-1}\tanh[\hat{E}/(2k_BT)], \tag{28}$$

where the the order parameter matrix $\hat{\Delta}$ is defined by its matrix elements Δ_{ij} [Eq. (19)].

Equations (27), (28), and (19) constitute self-consistent equations for the equilibrium $\hat{\rho}$, $\hat{\sigma}$, and $\hat{\Delta}$. The $\hat{E}$ matrix is defined as

$$\hat{E}=\sqrt{\hat{\Delta}^2+\hat{\varepsilon}^2}, \tag{29}$$

where matrix elements of $\hat{\varepsilon}$ in the single-electron basis set are

$$\varepsilon_{ij}\equiv t_{ij}+\sum_{kl}\ [\tilde{V}_{iljk}-(1/2)\tilde{V}_{ilkj}]\rho_{kl} \tag{30}$$

and

$$\tilde{V}_{iklm}\equiv\frac{1}{2}[V_{iklm}+V_{kiml}]. \tag{31}$$

Substituting Eqs. (27), (28), and (19) into Eq. (17) gives

$$\mathcal{H}(\hat{\rho},\hat{\sigma},\mu,T)=\mathrm{tr}\left[2\hat{E}\hat{n}+\hat{\varepsilon}-\hat{E}+\hat{\Delta}\hat{\sigma}-\frac{1}{2}\hat{M}\hat{\rho}\right], \tag{32}$$

where the matrix elements of $\hat{M}$,

$$M_{ij}=\sum_{kl}\ \left[\tilde{V}_{iklj}-\frac{1}{2}\tilde{V}_{ikjl}\right]\rho_{kl}, \tag{33}$$

and the $\hat{n}$ matrix is

$$\hat{n}\equiv[\exp(\hat{E}/2k_BT)+\mathbb{1}]^{-1}. \tag{34}$$

At zero temperature Eq. (32) gives the ground-state energy.

III. EQUATIONS OF MOTION FOR GENERALIZED COHERENT STATES

We are interested in the dynamics of the system coupled to two external fields: $v_s(\mathbf{r})$, which is coupled to the normal density $\rho(\mathbf{r})=\rho(\mathbf{r},\mathbf{r})$, and $\Delta_{ext}(\mathbf{r},\mathbf{r}')$, which is coupled to the anomalous density matrix $\sigma(\mathbf{r},\mathbf{r}')$.[10,9] These fields account for exchange correlation and provide a starting point for the TDDFT framework.[9] The total Hamiltonian then becomes $\hat{H}_T=\hat{H}+\hat{H}_{ext}$, where $\hat{H}$ is given by Eq. (3) and $\hat{H}_{ext}$ represents the interaction with the fields[9,10]:

$$\begin{aligned}\hat{H}_{ext}=&\sum_{\nu}\ \int\ d\mathbf{r}v_{ext}(\mathbf{r},t)\hat{\psi}^\dagger_\nu(\mathbf{r})\hat{\psi}_\nu(\mathbf{r})\\ &-\sum_{\nu\nu',\nu\neq\nu'}\ \int\ d\mathbf{r}\int\ d\mathbf{r}'\Delta_{ext}(\mathbf{r},\mathbf{r}',t)\hat{\psi}^\dagger_\nu(\mathbf{r})\hat{\psi}^\dagger_{\nu'}(\mathbf{r}')\\ &-\sum_{\nu\nu',\nu\neq\nu'}\ \int\ d\mathbf{r}\int\ d\mathbf{r}'\Delta^*_{ext}(\mathbf{r},\mathbf{r}',t)\hat{\psi}_\nu(\mathbf{r})\hat{\psi}_{\nu'}(\mathbf{r}').\end{aligned} \tag{35}$$

Our goal is to compute the dynamics of the system given by the total Hamiltonian $\hat{H}_T$ with the time-dependent external fields using the GCS ansatz for the many-electron density matrix, Eq. (8). This can be accomplished by applying the closed equations of motion for the averages of GCS generators [see Eq. (A9) in Appendix A]. These equations were obtained from the finite-temperature time-dependent variational principle.[17,18,30] Since $\rho_{i\nu j\nu}$, $\sigma^*_{i\nu j-\nu}$, and $\sigma^*_{i\nu j-\nu}$ are averages of GCS generators, we can immediately derive closed variational equations of motion for these quantities in real space.[17,18] Substituting the parameters of the energy $\mathcal{H}$ [Eq. (17)] and $\hat{H}_{ext}$ [Eq. (35)] in the closed equations of motion for the averages of GCS generators, Eq. (A9), we get ($\hbar=1$)

$$i\frac{\partial\rho(\mathbf{r},\mathbf{r}',t)}{\partial t}=-\frac{1}{2m_b}(\nabla_{\mathbf{r}}^2-\nabla_{\mathbf{r}'}^2)\rho(\mathbf{r},\mathbf{r}',t)+\int d\mathbf{r}_2[V(\mathbf{r}-\mathbf{r}_2)-V(\mathbf{r}'-\mathbf{r}_2)][\rho(\mathbf{r},\mathbf{r}',t)\rho(\mathbf{r}_2,\mathbf{r}_2,t)-\rho(\mathbf{r},\mathbf{r}_2,t)\rho(\mathbf{r}_2,\mathbf{r}',t)]$$
$$-\int d\mathbf{r}_2[W(\mathbf{r}-\mathbf{r}_2)-W(\mathbf{r}'-\mathbf{r}_2)][\sigma^*(\mathbf{r},\mathbf{r}_2,t)\sigma(\mathbf{r}_2,\mathbf{r}',t)-\sigma(\mathbf{r},\mathbf{r}_2,t)\sigma^*(\mathbf{r}_2,\mathbf{r}',t)]$$
$$+[v_{ext}(\mathbf{r},t)-v_{ext}(\mathbf{r}',t)]\rho(\mathbf{r},\mathbf{r}',t), \tag{36}$$

$$i\frac{\partial\sigma^*(\mathbf{r},\mathbf{r}',t)}{\partial t}=-\frac{1}{2m_b}(\nabla_{\mathbf{r}}^2-\nabla_{\mathbf{r}'}^2)\sigma^*(\mathbf{r},\mathbf{r}',t)+\int d\mathbf{r}_2[V(\mathbf{r}-\mathbf{r}_2)-V(\mathbf{r}'-\mathbf{r}_2)][\sigma^*(\mathbf{r},\mathbf{r}',t)\rho(\mathbf{r}_2,\mathbf{r}_2,t)-\sigma^*(\mathbf{r},\mathbf{r}_2,t)\rho(\mathbf{r}_2,\mathbf{r}',t)]$$
$$-\int d\mathbf{r}_2[W(\mathbf{r}-\mathbf{r}_2)-W(\mathbf{r}'-\mathbf{r}_2)][\sigma^*(\mathbf{r},\mathbf{r}_2,t)\rho(\mathbf{r}_2,\mathbf{r}',t)-\rho(\mathbf{r},\mathbf{r}_2,t)\sigma^*(\mathbf{r}_2,\mathbf{r}',t)]$$
$$+[\Delta_{ext}(\mathbf{r},\mathbf{r}',t)-\Delta_{ext}(\mathbf{r},\mathbf{r}',t)]\sigma^*(\mathbf{r},\mathbf{r}',t). \tag{37}$$

The order parameter $\Delta(\mathbf{r},\mathbf{r}',t)$, which characterizes the excitation gap in the spectrum [Eq. (29)], is defined as

$$\Delta(\mathbf{r},\mathbf{r}',t)\equiv\int d\mathbf{r}_2 W(\mathbf{r}-\mathbf{r}_2)\sigma(\mathbf{r}_2,\mathbf{r}',t). \tag{38}$$

It is interesting to note that the stationary solution of Eqs. (36) and (37) gives the normal $\rho(\mathbf{r},\mathbf{r}')$ and anomalous $\sigma(\mathbf{r},\mathbf{r}')$ density matrices, which minimize the equilibrium free energy, Eq. (11), in the ground state [Eqs. (27) and (28)].[17] The calculation of the free energy in Sec. II is thus not necessary for the present derivation. The formalism of the Appendix A) allows us to proceed directly from the ansatz, Eq. (8), to Eqs. (36) and (37). The calculations of Sec. II provide a consistency check and connect our results with more conventional derivations.

DFT is usually formulated in Hilbert space and involves the solution of four self-consistent equations for the Bogoliubov transformation coefficients $u(\mathbf{r})$ and $v(\mathbf{r})$, and the charge density $n(\mathbf{r})=\rho(\mathbf{r},\mathbf{r})$, and the anomalous density matrix $\sigma(\mathbf{r},\mathbf{r}')$ [Eqs. (4.121), (4.129), and (4.130) in Ref. 8]. In the presence of the external time-dependent field, the ordinary TDDFT framework involves also the solution of a system of four self-consistent equations for Bogoliubov transformation coefficients $u(\mathbf{r},t)$ and $v(\mathbf{r},t)$, the local anomalous density $\sigma(\mathbf{r},t)$, and the current density $\mathbf{j}(\mathbf{r},t)$ [Eqs. (20), (21), and (22) in Ref. 9]. Here, in contrast, we obtain the ground-state free energy and density matrices $\rho(\mathbf{r},\mathbf{r}',t)$ and $\sigma(\mathbf{r},\mathbf{r}',t)$ by the stationary solution of two equations, Eqs. (36) and (37). The solution of these equations gives the charge density $n(\mathbf{r},t)\equiv\rho(\mathbf{r},\mathbf{r},t)$ and the anomalous density matrix $\sigma(\mathbf{r},\mathbf{r}',t)$.

Equations (36) and (37) unify several widely used equations: if we neglect the pairing attraction $W=0$ and set $\sigma(\mathbf{r},\mathbf{r}',t)=\sigma^*(\mathbf{r},\mathbf{r}',t)=\Delta(\mathbf{r},\mathbf{r}',t)=0$, the last integral on the right-hand side (RHS) of Eq. (36) vanishes, and Eq. (36) reduces to the standard RPA equation.[27] By neglecting the Coulomb repulsion $V=0$, we obtain the BCS equations[1] [where on the RHS of Eqs. (36) and (37) the first integrals vanish]. Neglecting the second term on the RHS of Eq. (37) gives Eliashberg's equations,[2] which incorporate the Coulomb repulsion between electrons at the mean-field level.

Since our ansatz for the many-electron density matrix, Eq. (8), and the ansatz for the wave function, Eq. (7), have the same number of variational parameters h and Δ, they yield the same equations of motion for the averages of the GCS generators (i.e., normal and anomalous single-electron density matrices). The only dependence on temperature and chemical potential is through the initial conditions [Eqs. (27) and (28)]. Equations (36) and (37) conserve the temperature and chemical potential at all times.

In analogy with the RPA analysis,[27] we can look for a solution for the density matrices in the form of the following equations for matrices:

$$\hat{\rho}=\alpha\hat{X}\exp(-i\hat{\omega}t)+\alpha^*\hat{Y}^*\exp(i\hat{\omega}^*t),$$
$$\hat{\sigma}=\alpha\hat{\tilde{X}}\exp(-i\hat{\omega}t)+\alpha^*\hat{\tilde{Y}}^*\exp(i\hat{\omega}^*t). \tag{39}$$

Substituting Eq. (39) into Eqs. (36) and (37), we obtain the spectrum of the collective excitations:

$$\hat{\omega}=\sqrt{\hat{\Delta}^2+\hat{\varepsilon}^2}, \tag{40}$$

where ε_{kj} and Δ_{kj} are given by Eqs. (30) and (28), respectively.

IV. APPLICATION TO THE KOSTERLITZ-THOULESS PHASE TRANSITION

Recent studies of high-temperature superconductors[4,31,32] show a competition between two types of interaction between electrons: a pairing attraction, which makes the spectrum satisfy the Landau criterium of superfluidity by creating a gap in the excitation spectrum,[1] and Coulomb repulsion, which tends to eliminate the gap, thereby destroying the superconductivity. This competition leads to ''stripe'' high-temperature superconductivity (HTS) in cuprates [e.g., $Yba_2Cu_3O_{7-\delta}$ (YBCO)] above the low-oxygen-concentration threshold ($\delta\sim 0.20$).

When $\Delta(\mathbf{r},\mathbf{r}',t)\neq 0$ the excitation spectrum satisfies the Landau criterium of superfluidity, and a 3D system

becomes superconducting. However, cuprates (YBCO) are 2D structures.[4,5] The Kosterlitz-Thouless transition temperature[6,7] to the superfluid state in a two-dimensional superconductive system is given by $T_c=[\pi\hbar^2 n_s(T_c)]/(4k_B m_b)$,[6,7] where $n_s(T_c)$ is the temperature-dependent superfluid density of the superconductive system and k_B is the Boltzmann constant. For temperatures close to the phase transition in the mean-field approximation $[\Delta_{00}(T_c^0)=0]$, which satisfy $T-T_c^0 \ll T_c^0$, the superfluid density is $n_s(T)=[2(T_c^0-T)n_{2D}]/(T_c^0)$,[33] where $n_{2D}=p_F^2/(2\pi\hbar^2)$ is the total 2D density of electrons (p_F is a Fermi radius). To find T_c^0 from the condition $\Delta_{00}(T_c^0)=0$ one needs to solve the self-consistent equations (28) and (29) to obtain the temperature dependence of the order parameter $\Delta(T)$. The temperature of the phase transition can be estimated in the mean-field approximation.[33] For the gap spectrum of collective excitations, Eq. (40), the mean-field transition temperature T_c^0 is $\Delta_{00}=1.76k_B T_c^0$,[33] where Δ_{00} is the order parameter at zero temperature. Combining these expressions, we obtain for the temperature of the Kosterlitz-Thouless transition, below which the superconductivity exists,

$$T_c=\left(\frac{2k_B m_b}{\pi\hbar^2 n_{2D}}+\frac{1.76k_B}{\Delta_{00}}\right)^{-1}. \tag{41}$$

T_c can be calculated using the order parameter obtained by solving Eq. (28).

Since both T_c [Eq. (41)] and the order parameter Δ [Eq. (28)] decrease with Coulomb electron-electron repulsion V [Eq. (5)] and increase with electron-electron attraction W, Eq. (6), Eqs. (41) and (28) allow us to study the interplay of Coulomb repulsion and pairing attraction between electrons in FLSBS.[4] Cuprates have 2D structure,[4,5] where the electron-electron Coulomb correlations may not be neglected.[34,35] The screened 2D long-range Coulomb potential[36] in momentum space is $V_{2D}(p)=(2\pi e^2)/(p+\kappa_{2D})$, where the 2D Thomas-Fermi screening radius is density independent, $\kappa_{2D}=\hbar^2/(2m_b e^2)$. While the 3D short-range Coulomb potential in momentum space, $V_{3D}(p)=(4\pi e^2)/(p^2+\kappa_{3D}^2)$, where the 3D Thomas-Fermi screening radius[37] increases with density $\kappa_{3D}\sim n_{3D}^{1/3}$ and almost eliminates the Coulomb potential at large distances ($r>\kappa_{3D}^{-1}$). Therefore, the Coulomb electron-electron correlations in Eq. (28) are much more important in the 2D order parameter compared to 3D. The present theory thus describes the contribution of these correlations to the spectrum of collective excitations, the order parameter, superfluid density, the temperature of the Kosterlitz-Thouless transition, and the DFT exchange-correlation potential.

V. DISCUSSION

Using the GCS ansatz [Eq. (8)] for the many-electron density matrix we have calculated the ground-state free energy, the equations of motion for the normal and anomalous density matrices, and the quasiparticle spectrum of superconductors. This results in a quadratic expression for the energy, Eq. (32), in the operators $\hat{a}_i$ and $\hat{a}_i^\dagger$, without using the Bogoliubov algebra of $u-v$ transformations.[1]

The advantage of the Liouville space representation of TDDFT for the normal and anomalous density matrices is that it only requires one to solve two equations of motions for the normal and anomalous density matrices, Eqs. (36) and (37), coupled to two artificial external fields $v_{ext}(\mathbf{r})$ and $\Delta_{ext}(\mathbf{r},\mathbf{r}')$, which contain exchange correlation,[8] instead of the four self-consistent equations for Bogoliubov coefficients u and v and density matrices ρ and σ. The normal $\rho(\mathbf{r},\mathbf{r}')$ and anomalous $\sigma(\mathbf{r},\mathbf{r}')$ density matrices, which minimize the equilibrium free energy Eq. (11) (Ref. 17) in the ground state, are simply given by the stationary solution of the Eqs. (36) and (37). And in order to get this ground state we actually do not need to derive the parameters of the effective quadratic Hamiltonian, Eq. (9), for the many-electron density matrix, as we did in Sec. II.

Our equations for the total energy, the spectrum of collective excitations, and the gap are written in a general basis set and therefore apply to both homogeneous and nonhomogeneous systems. In a homogeneous system we can use the plane-wave basis; i.e., for the two-dimensional system the eigenfunctions of a momentum $\mathbf{p}$, $\phi_{\mathbf{p}}(\mathbf{r})=U^{-1/2}\exp(-i\mathbf{p}\cdot\mathbf{r})$, where U is the volume. Since we used a general basis set (not necessarily plane waves), our results, which contain nonuniform normal $\rho(\mathbf{r},\mathbf{r}')$ and anomalous $\sigma(\mathbf{r},\mathbf{r}')$ density matrices, should be able to describe the short-coherence-length superconductors: for example, in YBCO, where the 1-nm coherence length is comparable to the lattice constant.[4]

Finally, we comment on the connection of our results to the ground- state energies and the collective spectrum of excitations in superconductors calculated using density-functional theory and time-dependent density functional theory.[8–11] Let us consider the following exchange-correlation potentials that depend on the density matrix (rather than merely on the charge density):

$$v_{xc}[\rho](\mathbf{r})=-\frac{1}{2}\frac{\delta}{\delta n(\mathbf{r})}\times\left[\int d\mathbf{r}\int d\mathbf{r}'\frac{\rho(\mathbf{r},\mathbf{r}')\rho(\mathbf{r}',\mathbf{r})}{|\mathbf{r}-\mathbf{r}'|}\right]\Bigg|_{n(\mathbf{r}')=\overline{\rho}(\mathbf{r}',\mathbf{r}')}. \tag{42}$$

Substituting Eq. (42) into the DFT equations for the charge density and the anomalous density matrix[8,10] gives Eqs. (27) and (28) for the normal and anomalous density matrices. The GCS ansatz [Eq. (7)] is thus equivalent to TDDFT provided we use the approximate exchange-correlation potential, Eq. (42). To improve this functional, the adiabatic (time-independent) exchange-correlation potentials can be obtained using the functional derivatives[8]

$$v_{xc}([\rho,\sigma];\mathbf{r})=\frac{\delta F_{xc}[\rho,\sigma]}{\delta\rho(\mathbf{r})},$$

$$\Delta_{xc}([\rho,\sigma];\mathbf{r},\mathbf{r}')=-\frac{\delta F_{xc}[\rho,\sigma]}{\delta\sigma^*(\mathbf{r},\mathbf{r}')}, \tag{43}$$

where the exchange-correlation free energy F_{xc} can be obtained using Feynmann diagrammatic perturbation theory for the self-energy of the Green function.[21,26] The zero-order normal and anomalous density matrices derived using the present ansatz [Eq. (7)] are given by Eqs. (36) and (37), respectively. The spectrum of the corresponding Green function is E_{ij} given by Eq. (29) together with Eq. (30).

The first-order exchange-correlation contribution to the order parameter is identical to that of Ref. 26 for homogeneous superconductors provided we set $\varepsilon_{ij}=t_{ij}$. The second term appearing in our expression for the energy ε_{ij}, Eq. (30),

$$\sum_{kl}\left[\tilde{V}_{iljk}-\frac{1}{2}\tilde{V}_{ilkj}\right]\rho_{kl},$$

comes from the first term $\alpha_{ij}(t)\hat{a}^{\dagger}_{i\nu}\hat{a}_{j\nu'}$ in the exponential in our ansatz, Eq. (7), which represents electron-electron exchange. It corrects each order in the perturbative series of Ref. 26 for the exchange-correlation potential. This term is absent in the OKG equations which use as a reference the Bogoliubov–de Gennes approximation, which takes into account the BCS pairing and Coulomb correlations at the Hartree level[12] and neglects exchange-correlation potentials.[9,10]

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APPENDIX: GENERALIZED COHERENT-STATE REPRESENTATION OF A HAMILTONIAN WITH ELECTRON-ELECTRON PAIRING

Mathematically, a set of GCS's is determined by a Lie group G, its irreducible unitary vector representation T with the space V, and a reference state $|\Omega\rangle\in V$. The GCS's are then states that have a form $T(g)|\Omega\rangle$ with $g\in G$, where g is a set of parameters.[16–18]

Our ansatz for the many-electron density matrix [Eq. (8)] can be expressed as

$$K(t)=\frac{1}{Z}\exp\left(\sum_i \lambda_i\hat{T}_i\right), \tag{A1}$$

where the set of numbers λ_i parametrizes the density matrix. The operator set $\{\hat{T}_i\}=\{\hat{a}^{\dagger}_{i\pm\nu}\hat{a}_{j\pm\nu},\hat{a}^{\dagger}_{i\nu}\hat{a}^{\dagger}_{j-\nu},\hat{a}_{i\nu}\hat{a}_{j-\nu},\hat{I}\}$ ($\hat{I}$ is the identity operator) which forms the Lie group G is characterized by the commutation relations among the complete set of operators $\hat{T}_i$ necessary for describing the quantum dynamics of the system:

$$[\hat{T}_i,\hat{T}_j]=\sum_k C^k_{ij}\hat{T}_k, \tag{A2}$$

where C^k_{ij} are known as the structure constants of the set $\{\hat{T}_i\}$. Writing $\hat{T}^{(-)}_{ij}\equiv\hat{a}_{i\nu}\hat{a}_{j-\nu}$, $\hat{T}^{(+)}_{ij}\equiv\hat{a}^{\dagger}_{i\nu}\hat{a}^{\dagger}_{j-\nu}$, $\hat{T}^{(z)}_{ij}\equiv\hat{a}^{\dagger}_{i\pm\nu}\hat{a}_{j\pm\nu}+\frac{1}{2}\delta_{ij}\hat{I}$, and, since $\hat{a}_i$ satisfy Fermi anticommutation rules, we have the closed algebra of generators $\{\hat{T}_i\}$ with respect to the following commutation rules:

$$[\hat{T}^{(-)}_{ij},\hat{T}^{(-)}_{i'j'}]=[\hat{T}^{(+)}_{ij},\hat{T}^{(+)}_{i'j'}]=[\hat{T}^{(z)}_{ij},\hat{T}^{(z)}_{i'j'},]=0,$$

$$[\hat{T}^{(-)}_{ij},\hat{T}^{(+)}_{i'j'}]=\delta_{i-ji'-j'}(1-(\hat{T}^{(z)}_{ii}+\hat{T}^{(z)}_{-i-i})),$$

$$[\hat{T}^{(z)}_{ij},\hat{T}^{(-)}_{i'j'}]=-\delta_{ii'}\delta_{j-j'}\hat{T}^{(-)}_{j-j'}-\delta_{ij'}\delta_{j-i'}\hat{T}^{(-)}_{ji'},$$

$$[\hat{T}^{(z)}_{ij},\hat{T}^{(+)}_{i'j'}]=\delta_{ji'}\delta_{i-j'}\hat{T}^{(+)}_{ij'}+\delta_{jj'}\delta_{i-i'}\hat{T}^{(+)}_{ii'}. \tag{A3}$$

Equations (8), (A1), (A2), and (A3) show that our ansatz for the many-electron density matrix is generated by the operator set $\{\hat{T}_i\}$, which forms the closed algebra of generators with respect to their binary commutation. The states described by our ansatz, Eqs. (8), thus constitute generalized coherent states.

The set of generators $\{\hat{T}_i\}$ corresponds to the Hamiltonian, Eq. (1). The variational equations at zero temperature are derived as follows: given a Hamiltonian $\hat{H}$ and time-dependent wave functions $|\Omega(\tau)\rangle$, we minimize the action:

$$S[\Omega(\tau)]=\int d\tau[i\langle\Omega(\tau)|d\Omega(\tau)/d\tau\rangle-\langle\Omega(\tau)|\hat{H}|\Omega(\tau)\rangle]. \tag{A4}$$

By choosing $|\Omega(\tau)\rangle$ to be a GCS, the resulting variational equations can be written in the Hamiltonian form for any set of coordinates Ω_j which parametrize $|\Omega\rangle$:

$$\frac{d\Omega_j}{d\tau}=\{\mathcal{H},\Omega_j\}, \tag{A5}$$

where $\{\cdots\}$ denote Poisson brackets and $\mathcal{H}$ is the classical Hamiltonian defined by

$$\mathcal{H}(\Omega)=\langle\Omega|\hat{H}|\Omega\rangle. \tag{A6}$$

The Poisson brackets clearly establish the link between the variational equations and classical dynamics.

When the classical Hamiltonian is given by

$$\mathcal{H}=\sum_{n=1}^{k}\sum_{i_1\cdots i_n}h^{(n)}_{i_1\cdots i_n}\langle\hat{T}_{i_1}\rangle\cdots\langle\hat{T}_{i_n}\rangle, \tag{A7}$$

the Poisson brackets assume a very simple form provided the wave functions $|\Omega\rangle$ are parametrized by the expectation values $\langle\Omega|\hat{T}_j|\Omega\rangle$ of the operators $\hat{T}_j$ rather than by the parameters Ω_j. These expectation values then constitute a full set of parameters that uniquely specify the quantum state $|\Omega\rangle$. In particular, if $\hat{T}_j$ form a closed algebra, Eq. (A2), the Poisson brackets for $\langle\hat{T}_j\rangle$ are given by

$$\{\langle\hat{T}_m\rangle,\langle\hat{T}_n\rangle\}=i\sum_k C^k_{m,n}\langle\hat{T}_k\rangle, \tag{A8}$$

and the variational equations of motion for $\langle\hat{T}_m\rangle$ take the closed form

$$i\frac{d\langle\hat{T}_m\rangle}{d\tau}=\sum_{n=1}^{k}\sum_{j=1}^{n}\sum_{i_0\cdots i_n}C^k_{mi_0,\cdots,i_j}h^{(n)}_{i_1\cdots i_n}\langle\hat{T}_{i_1}\rangle\cdots\langle\hat{T}_{i_n}\rangle. \tag{A9}$$

Using Eq. (7), it therefore suffices to derive the equations of motion for the expectation values $\langle\hat{a}_i^\dagger\hat{a}_j\rangle$ and $\langle\hat{a}_i\hat{a}_j\rangle$ to uniquely specify the dynamics of electrons. This may be done using the differential property of the Poisson brackets:

$$\{f,gh\}=-\{gh,f\}=\{f,g\}h+g\{f,h\}. \tag{A10}$$

When the expectation values are for generators of the set of GCS's of some Lie group G, their Poisson brackets are given by the commutators of the underlying generators of the group. This direct correspondence between ordinary quantum mechanical commutators and the Poisson brackets greatly simplifies the calculation, since the variational procedure is then equivalent to the Heisenberg equations of motion. Equations. (36) and (37) were obtained using Eq. (A9).

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