Superoperator coupled cluster method for nonequilibrium density matrix

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Abstract
We develop a superoperator coupled cluster method for nonequilibrium open many-body quantum systems described by the Lindblad master equation. The method is universal and applicable to systems of interacting fermions, bosons or their mixtures. We present a general theory and consider its application to the problem of quantum transport through the system with electron–phonon correlations. The results are assessed against the perturbation theory and nonequilibrium configuration interaction theory calculations.

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(Some figures may appear in colour only in the online journal)

1. Introduction
The coupled cluster method has proved to be extremely useful in a wide variety of many-body calculations ranging from nuclear physics to quantum chemistry [1–3]. In this method, the wave function of a quantum many-body system is decomposed in terms of amplitudes for excited ‘clusters’ of a finite number of quasiparticles or particle-hole pairs. Coupled cluster theory works for systems of both bosons and fermions and gives high-precision results for the equilibrium properties. Today, it is one of the most widely used methods for equilibrium many-body calculations [3]. In this paper, we propose the extension of the coupled cluster method for nonequilibrium open quantum systems governed by the Lindblad master equation.

The paper is organized as follows. Section 2 describes the Liouville–Fock space and superoperator representation of the Lindblad master equation. Section 3 discusses the nonequilibrium coupled cluster (NECC) expansion for the steady-state density matrix of a
correlated many-body system. In section 4, we apply the method to the problem of charge transport through the system with electron–phonon interaction and compare the numerical results with other methods. Section 5 summarizes the main results of the paper. Appendix A contains some technical details concerning the choice of a reference state for transport calculations. In appendix B, we give explicit equations for the cluster amplitudes.

2. Lindblad equation in Liouville–Fock space

We consider a correlated quantum system described by the Hamiltonian $H$. The system is open and exchanges energy and particles with the environment. We assume that the reduced density matrix of the system satisfies the Lindblad master equation [4]:

$$i \frac{\partial \rho(t)}{\partial t} = [H, \rho(t)] + i \Pi \rho(t),$$  \hspace{1cm} (1)

where $\Pi \rho(t)$ is the non-Hermitian dissipator given by the standard Lindblad form

$$\Pi \rho(t) = \sum_k \left( 2L_k \rho(t)L_k^\dagger - \left( L_k^\dagger L_k, \rho(t) \right) \right),$$  \hspace{1cm} (2)

and $L_k$ are so-called Lindblad operators, which model the effects of the environment [4]. In what follows, we consider that $H$ and $L_k$ act in the Fock space of the system under consideration, so they can be written in terms of creation and annihilation operators. The Lindblad master equation (1) describes the time evolution of an open system preserving Hermiticity, normalization and positivity of the reduced density matrix.

The Lindblad master equation can be converted to a non-Hermitian Schrödinger-like form with the use of superoperator formalism [5–8]. Within the formalism, every Fock space operator $A$ is considered as a super-vector $|A\rangle$ in the Liouville–Fock space. In particular, if $|m\rangle$ and $|n\rangle$ are vectors in the Fock space, then $|mn\rangle \equiv |m\rangle |n\rangle$ is a super-vector in the Liouville–Fock space. The scalar product in the Liouville–Fock space is defined as $\langle A | T_r[A]^{1\dagger} 1 | B \rangle = \text{Tr}[A^\dagger B]$. In the Liouville–Fock space, we can introduce creation and annihilation superoperators $\hat{a}^\dagger$, $\hat{a}^\dagger$ and $\hat{\alpha}$, $\hat{\alpha}$:

$$\hat{a}_k |mn\rangle = |a_k [m](n)\rangle, \quad \hat{\alpha}_k |mn\rangle = \tau_{mn} |m\rangle |n\rangle \hat{a}_k^\dagger,$$

$$\hat{\alpha}_k^\dagger |mn\rangle = |a_k^\dagger [m](n)\rangle, \quad \hat{\alpha}_k^\dagger |mn\rangle = \tau_{mn} |m\rangle |n\rangle \hat{a}_k,$$  \hspace{1cm} (3)

where $|m\rangle = |m_1, m_2, \ldots \rangle$ are eigenvectors of the particle number operator, i.e., $a_k^\dagger a_k |m\rangle = m_k |m\rangle$. The phase $\tau_{mn} = 1$ for bosonic creation and annihilation superoperators, while for fermionic ones we have $\tau_{mn} = i(-1)^\mu$, where $\mu = \sum_k (m_k + n_k)^\dagger$. For an operator in the Fock space $A = A(a^\dagger, a)$, we formally define two superoperators $\hat{A} = A(\hat{a}^\dagger, \hat{a})$, $\hat{A} = A^\dagger(\hat{a}^\dagger, \hat{\alpha})$ and refer to them as nontilde and tilde superoperators, respectively. Then, we can find that the respective super-vector $|A\rangle$ is given by

$$|A\rangle = \hat{A} |I\rangle = \sigma_a \hat{A}^\dagger |I\rangle,$$  \hspace{1cm} (4)

where the supervector $|I\rangle$ corresponds to the Fock space identity operator, and the phase $\sigma_a = -i(\pm 1)$ if $A$ is a fermionic (bosonic) operator. Moreover, for the product of operators,

3 Such defined creation and annihilation superoperators are Hermitian conjugate to each other, $\hat{a}^\dagger = (\hat{a})^\dagger$, $\hat{a}^\dagger = (\hat{a})^\dagger$, and satisfy the same (anti)commutation relations as their Fock space counterparts, $[\hat{a}, \hat{a}^\dagger]_\mathbb{C} = 1$ etc. See [8] for more details.
the following relations hold:
\[ \langle A_1 A_2 \rangle = \hat{A}_1 \langle A_2 \rangle = \tau \tilde{A}_2 \langle A_1 \rangle. \] (5)

Here, \( \tau = i \) if both \( A_1 \) and \( A_2 \) are fermionic and \( \tau = \sigma_{A_2} \) otherwise. The Liouville–Fock space is the physical name for what is known in mathematics as the CAR/CCR algebra of observables, which admits, the Gelfand–Naimark–Segal (GNS) representation, and the Liouville superoperator defined below is the generator of dynamics in this larger space.

Using the correspondence between Fock space operators and Liouville–Fock space super-vectors and relations (5), we rewrite the Lindblad master equation (1) as a Schrödinger-like equation for the density matrix
\[ i \frac{\partial}{\partial t} \langle \rho(t) \rangle = L \langle \rho(t) \rangle. \] (6)

Here, the superoperator \( L \) (Liouvillian) is given by
\[ L = \hat{H} - \bar{H} - i \sum_k \Pi_k, \] (7)

and the non-Hermitian dissipators \( \Pi_k \) read as
\[ \Pi_k = L_k^\dagger L_k + \bar{L}_k^\dagger \bar{L}_k - 2\sigma_{L_k} \bar{L}_k^\dagger L_k. \] (8)

The statistical average of any operator is
\[ \langle A \rangle = \text{Tr} \left[ L |A \rangle \langle \rho(t) | \right] = \langle L \hat{A} | \langle \rho(t) \rangle \rangle = \langle L \hat{A} | \langle \rho(t) \rangle \rangle. \] (9)

Before proceeding further, let us mention some important properties of the Liouvillian and introduce terminology that will be used throughout the paper. The connection between nontilde and tilde superoperators is given by the tilde-conjugation rules
\[ \left( c_1 \hat{A}_1 + c_2 \hat{A}_2 \right) \sim = c_1^* \tilde{A}_1 + c_2^* \tilde{A}_2, \quad \left( \hat{A}_1 \hat{A}_2 \right) \sim = \tilde{A}_1 \tilde{A}_2, \quad \left( \tilde{A} \right) \sim = \hat{A}. \] (10)

By applying tilde-conjugation to equation (6), we get \( \langle L \rangle \sim = -L \). The super-vector is called tilde-invariant if \( \langle A \rangle \sim \equiv \tilde{\hat{A}} | \langle \rangle = \langle A \rangle \). The examples of tilde-invariant super-vectors are \( |l \rangle \) and \( |\rho(t) \rangle \). Taking the time derivative of the normalization condition
\[ \langle l | \rho(t) \rangle = \text{Tr} \left[ |\rho(t) \rangle \langle \rho(t) | \right] = 1, \] (11)

we find \( \langle l | L = 0 \), i.e., \( l \) is the left zero-eigenvalue eigenvector of \( L \).

**3. Coupled cluster expansion of density matrix**

Consider the problem of obtaining the steady-state solution of equation (6). We wish to find the density matrix, which corresponds to the right zero-eigenvalue eigenstate of the Liouvillian
\[ L |\rho \rangle = 0. \] (12)

The problem is much more complicated than in the equilibrium case, since the Liouvillian \( L \) is non-Hermitian and its spectrum is not bound from below, so that the zero-eigenvalue eigenvector can not be found variationally.

If the system is quasi-free, i.e., there are no two-body interactions, the Liouvillian is a non-Hermitian quadratic form that can be readily diagonalized by nonunitary, canonical transformations:
Here, $\hat{\xi}$, $\hat{\bar{\xi}}$ describe the normal modes of quadratic Liouvillian (so-called nonequilibrium quasiparticles). The creation and annihilation superoperators for nonequilibrium quasiparticles are not mutually Hermitian conjugates but, nevertheless, they satisfy canonical (anti) commutation relations [7, 8]. With the use of nonequilibrium quasiparticles, the uncorrelated steady-state density matrix

$$L_0 \left\langle \rho_0 \right\rangle = 0$$

(14)
can be defined as a vacuum of nonequilibrium quasiparticles: $\hat{\xi}_n \left\langle \rho_0 \right\rangle = \hat{\bar{\xi}}_n \left\langle \rho_0 \right\rangle = 0$. Note that the bra super-vector $\langle I |$ plays the role of the right vacuum, i.e., $\langle I | \hat{\xi}_n \rangle = \langle I | \hat{\bar{\xi}}_n \rangle = 0$. Besides, the right and left vacuums are normalized as $\langle I | \rho_0 \rangle = 1$.

We now consider how correlations induced by interaction between nonequilibrium quasiparticles modify this steady-state vacuum. Likewise to the equilibrium case, where correlations produce particle-hole or quasiparticle excitations over an uncorrelated ground state, the correlations produce nonequilibrium multiquasiparticle excitations over the reference state $\langle I | \rho_0 \rangle$. By analogy with the coupled cluster approach widely used in equilibrium many-body calculations, we propose an exponential ansatz for the correlated nonequilibrium steady-state density matrix

$$\rho = e^S \left\langle \rho_0 \right\rangle,$$

(15)

where the so-called cluster correlation superoperator $S$ is constructed as a linear combination of multiconfigurational creation superoperators:

$$S = \sum_i s_i C_i^+, \quad (16)$$

and $C_i^+$ is a product of creation superoperators $\hat{\xi}_i^+, \hat{\bar{\xi}}_i^+$. The expansion coefficients $s_i$ are called cluster amplitudes. Because the density matrix is tilde invariant, the cluster correlation superoperator $S$ should be invariant with respect to tilde-conjugation, i.e., $(S) = S$. Also note that from $\langle I | C_i^+ \rangle = 0$ it follows that the normalization condition (11) is automatically fulfilled for the ansatz.

With the exponential ansatz for the density matrix, the exact equation (12) is then rewritten in the similarity-transformed form

$$e^{-S} L e^S \left\langle \rho_0 \right\rangle = 0.$$

(17)

By taking a scalar product of this equation with the complete set of states $\langle I | C_j \rangle$, $\forall j$ and applying the nested commutator expansion, we obtain a set of coupled multinomial equations for the cluster amplitudes $s_j$:

$$\langle I | C_j \left\{ L + [L, S] + \frac{1}{2!} [[L, S], S] + \ldots \right\} \left\langle \rho_0 \right\rangle = 0, \quad \forall j. \quad (18)$$

Performing the commutations leads to contractions between creation and annihilation superoperators. Considering that $C_j^+$ are formed by creation superoperators, the only possible contractions occur between annihilation superoperators in the Liouvillian $L$ and creation superoperators in $S$. Following equilibrium coupled cluster theory we call each such contraction as a ‘link’ [2]. Thus, we can say that each element of $S$ in parametrization (16) is linked or coupled directly to $L$. Moreover, since $L$ contains a finite number of annihilation superoperators and each commutator removes one annihilation superoperator, the infinite
series of commutators in equation (18) terminates at a finite order. As a result, we obtain finite-order polynomial equations for the cluster amplitudes \( s_i \). Furthermore, since \( S \) is \( \tilde{\text{g}} \)-invariant, we have \( s_i = s_i^\dagger \) for \( C_i^\dagger = (C_i^\dagger)^\dagger \).

The NECC method would be exact if all possible multiconfigurational creation superoperators were included in the expansion (16). In any real many-body calculations this is usually impossible to achieve, and we should truncate the expansion (16) at some lower order. As a result, we get an approximate nonequilibrium steady-state density matrix.

At the end of this section, we note that there is a certain degree of flexibility in defining the uncorrelated reference state \( |\rho_0\rangle \) for subsequent coupled cluster expansion. Depending on the problem, it is sometimes convenient to define normal modes \( \xi \) by diagonalizing only the part of \( L_0 \) and consider the rest of the \( L_0 \) as a perturbation that induces ‘correlations’ in the coupled cluster density matrix expansion.

4. Application of the nonequilibrium coupled cluster method: charge transport through a vibrational electronic level

4.1. Derivations of main equations

As an application of the NECC method, we consider the problem of charge transport through a correlated quantum system (central region) attached to two macroscopic electrodes, left (\( L \)) and right (\( R \)). The electrodes are modeled as infinite noninteracting electron reservoirs that have the same temperature \( T \) but different chemical potentials \( \mu_L, \mu_R \) through an applied voltage \( V = \mu_L - \mu_R \). The coupling between the central region and the electrodes is described by the tunneling Hamiltonian. For the central region, we consider one electronic level of energy \( \varepsilon_0 \) coupled linearly to a vibrational mode (phonon) of frequency \( \omega_0 \):

\[
H_0 = \varepsilon_0 \alpha^\dagger \alpha + \omega_0 d^\dagger d + \kappa \alpha^\dagger (d^\dagger + d),
\]

where \( \alpha^\dagger (\alpha) \) and \( d^\dagger (d) \) are electron and phonon creation (annihilation) operators, respectively. The problem is to compute the steady-state current through the central region.

In [8, 9] we have introduced the concept of embedding, which enables us to replace the finite system (central region + left and right electrodes) by a finite open system whose reduced density matrix is governed by the Lindblad master equation. The main idea is to divide each electrode into two parts: an infinite environment and a sufficiently large but finite buffer between the central region and the environment. Then, by projecting out into the environment degrees of freedom and employing the Born–Markov approximation, we obtain the Lindblad master equation (1) for the density matrix of the embedded system (central region + left and right buffers; see figure 1). The Hamiltonian of the embedded system has the form

\[
H = H_S + H_T + H_B,
\]

where \( H_B = \sum_{k \alpha} \varepsilon_{k\alpha} a^\dagger_{k\alpha} a_{k\alpha} \) describes the left (\( \alpha = L \)) and right (\( \alpha = R \)) finite buffers (\( k = 1, \ldots, N \)), and \( H_T = \sum_{k \alpha} l_{k\alpha} (a^\dagger_{k\alpha} \alpha + \text{h. c.}) \) is the tunneling coupling between the central region and the buffers. The non-Hermitian part of the master equation is

\[
\Pi \rho(t) = \sum_{k \alpha} \sum_{\mu=1,2} \left( 2L_{k\alpha\mu} \rho(t) L_{k\alpha\mu}^\dagger - \left\{ L_{k\alpha\mu}^\dagger L_{k\alpha\mu}, \rho(t) \right\} \right),
\]
where

\[ L_{k1} = \sqrt{\Gamma_{k1}} a_{k_1}, \quad L_{k2} = \sqrt{\Gamma_{k2}} \hat{a}_{k_2}. \]  

(22)

Here \( \Gamma_{k1} = \gamma_{k1} (1 - f_{k1}) \), \( \Gamma_{k2} = \gamma_{k2} f_{k2} \), \( f_{k1} = [1 + e^{(\varepsilon_{k1} - \mu)/T}]^{-1} \) and \( \gamma_{k1} \) is determined through the imaginary part of the self-energy arising from the buffer-environment interaction.

Using the formalism of superoperators described in section 2, we rewrite the obtained master equation in the Schrödinger-like form (6). The Liouvillian of the embedded system is given by

\[ L = L_0 + L_T + L_\kappa. \]  

(23)

Here, \( L_0 = L_{el} + L_{ph} + L_B \) describes the uncorrelated system, \( L_T = \hat{H}_T - \bar{H}_T \), and

\[ L_\kappa = \kappa \left\{ \hat{a} \hat{\alpha} - \hat{\alpha} \hat{a} (t.c.) \right\}. \]  

(24)

represents the electron–phonon correlations. Hereinafter, the notation ‘(t.c.)’ stands for items which are tilde-conjugated (see the tilde-conjugation rules in section 2) to displayed ones. The steady-state current through the central region is given by

\[ J_R = i \sum_k t_{k1} \left\langle I \left| \hat{a}_{k1} \hat{\alpha} - \hat{\alpha} \hat{a}_{k1} \right| \rho \right\rangle. \]  

(25)

which requires the solution of equation (12) with Liouvillian (23).

To solve equation (12) within the NECC method, we should first define an appropriate reference state. For the problem under consideration, as the reference state we take the density matrix \( |\rho_0\rangle \) of the uncorrelated system, i.e.,

\[ L_0 |\rho_0\rangle = 0. \]  

(26)

In appendix A, we demonstrate how to introduce normal modes of \( L_0 \), such that \( (1 | \rho_0) \) would be the vacuum states for respective creation and annihilation superoperators. In terms of these uncorrelated superoperators, \( L_0 \) is diagonal.
\[ L_0 = \epsilon_0 \hat{\beta} \hat{\beta} + \omega_0 \hat{\gamma} \hat{\gamma} + \sum_{k \alpha} E_{k \alpha} \hat{b}_{k \alpha} \hat{b}_{k \alpha} - \text{(t.c.)}, \]  
while the parts of the Liouvillian responsible for electron–phonon correlations and the coupling between the central region and the buffers read as

\[ L_\varepsilon = \kappa \left\{ \hat{\beta} \hat{\beta} \left[ (1 + N_m) \hat{\gamma} + N_m \hat{\gamma} + \hat{\gamma} + \hat{\gamma} \right] + i \hat{\beta} \hat{\beta} \hat{\gamma} \right\} \]  
and

\[ L_T = -\sum_{k \alpha} I_{k \alpha} \left\{ \hat{\beta} \hat{b}_{k \alpha} + \hat{b}_{k \alpha} \hat{\beta} - i \hat{b}_{k \alpha} \hat{b}_{k \alpha} - \text{(t.c.)} \right\}, \]

respectively. In (28), \( N_m \) denotes the number of equilibrium thermally excited phonons (see appendix A).

Due to interaction terms (28, 29) the correlated steady-state density matrix contains multiconfigurational excitations above the reference state \( |\rho_0\rangle \). For the considered problem the multiconfigurational creation superoperators \( C_i^\dagger \) are a product of bosonic and fermionic creation superoperators. More specifically, the index \( i \) becomes the set of occupation numbers,

\[ n^i = (n^i_\beta, n^i_\beta, n^i_\beta, \ldots, n^i_\beta, n^i_\beta, n^i_\beta, \ldots), \]

and the generic multiconfigurational creation superoperators \( C_i^\dagger \) is given by

\[ C_i^\dagger \rightarrow \left( \hat{\gamma} \right)^{n^i_\gamma} \left( \hat{\beta} \right)^{n^i_\beta} \prod_{ak} \left( \hat{b}^\dagger_{ak} \right)^{n^i_{b_{ak}}} \prod_{ak} \left( \hat{b}^\dagger_{ak} \right)^{n^i_{b_{ak}}}. \]

Moreover, from the structure of \( L_\varepsilon \) and \( L_T \), it follows that in each \( C_i^\dagger \) the number of nontilde fermionic superoperators should be equal to the number of tilde ones, i.e.,

\[ n^i_\beta + \sum_{ak} n^i_{b_{ak}} = n^i_\beta + \sum_{ak} n^i_{b_{ak}}, \quad \forall i. \]

Due to the phonon subsystem, the number of possible multiconfigurational excitations is infinite. Furthermore, since \( L_\varepsilon \) involves terms with two annihilation superoperators, the expansion (18) terminates after the second-order commutators. Therefore, for the considered problem, the exact cluster superoperator \( S \) is given by the solution of the infinite system of coupled quadratic nonlinear equations.

In the current work, we restrict our consideration by the cluster correlation superoperators \( S_1 \) and \( S_2 \), which includes only terms linear with respect to phonon-creation superoperators. The superoperator \( S_1 \) is quadratic in terms of fermion creation superoperators

\[ S_1 = W \left( \hat{\gamma} + \hat{\gamma} \right) - i \hat{\gamma} \hat{\beta} \left( \hat{b}_k^\dagger + \hat{b}_k \right) \left( I_k + \hat{I}_{10} \hat{\gamma} + \hat{I}_{01} \hat{\gamma} \right) \]

\[ + \sum_k \left( \hat{b}^\dagger_k \hat{b}_k \right) \left( \hat{f}_{110} + \hat{f}_{010} \hat{\gamma} \right) \]

\[ + \sum_{kl} \left( \hat{b}^\dagger_k \hat{b}_l \right) \left( \hat{f}_{110} + \hat{f}_{010} \hat{\gamma} \right), \]

where...
while $S_2 = S_1 + S'$ contains four-fermion components

$$S' = + i \sum_{k,l} \hat{\beta}_k^\dagger \hat{\beta}_l^\dagger (G_{kl} + G_{kl01} \gamma^\dagger + G_{kl01} \gamma^\dagger)$$

Here, to simplify the notations, we omit the index $\alpha = L, R$ implying that the summation is performed over the states in the left and right buffers, i.e., $k, l \in L, R$. Substituting $S_1$ or $S_2$ into (18), we obtain the set of nonlinear (quadratic) equations for the cluster amplitudes $n, W, I, F$ and $G$ (see equation (B.1) in appendix B). The solution of these equations provides an approximate steady-state density matrix for the considered nonequilibrium charge transport problem. From $S_{1,2} = (S_{1,2})^\dagger$, it follows that $n$ and $W$ are real, while $n^* = n_{01}, F^\dagger = F_k, F_{kl0}^\dagger = F_{01k}, G^\dagger = G_k$, and $G_{kl01}^\dagger = G_{01k}$. In what follows, to distinguish between results obtained with $S_1$ and $S_2$ superoperators, we will refer to them as NECC(1) and NECC(2), respectively.

To compute the steady-state current, we express the current superoperator in equation (25) in terms of uncorrelated creation and annihilation superoperators. We get

$$J_\alpha = \sum_{k \in \alpha} l_k \left\{ I \left| \hat{\beta}_k \hat{\beta}_k^\dagger \right. \right\} = -2 \text{Im} \sum_{k \in \alpha} l_k I_k.$$  

Thus, the steady-state current is expressed through the cluster amplitudes $I_k$. On the other hand, with the above relation the first equation in (B.1) becomes

$$J_L + J_R = 0.$$  

Therefore, we can say that the first equation in (B.1) guaranties the current conservation. Moreover, it can be easily shown that this equation is exact in the sense that the inclusion of extra terms in $S_{1,2}$ does not modify it. Consequently, the presented coupled cluster approach for the charge transport problem is a current conserving one.

4.2. Numerical calculations

In our numerical calculations, the left and right electrodes are represented by two semi-infinite tight-binding chains of sites. The coupling between the central region and the edge sites is given by the matrix elements $t_{\alpha} (\alpha = L, R)$. Each electrode is characterized by the hopping matrix element $h_\alpha$ and the on-site energy $\epsilon_\alpha$. We choose the following parameters: $h_L = h_R = 2.5, t_L = t_R = 1.0$ and the temperature $T = 0.1$. Additionally, we assume that the electrodes are half filled, i.e., the corresponding left and right chemical potentials are positioned at $\epsilon_L, R$, and the applied voltage $V = 1.0$ symmetrically shifts the on-site energies, $\epsilon_{L,R} = \pm 0.5 V$.

For the embedded system, we take $N = 800$ atoms from each electrode as buffers. This size of the buffers has been proven to give the exact results for the steady-state current calculated within the mean-field approximation and the second-order perturbation (SOPT) theory [7, 8, 10]. In [11], we justify this choice of $N$ for nonequilibrium configuration interaction (NECI) calculations. Following [11], we studied the convergency of NECC results against increasing the size of buffers. We found that when increasing $N$, the NECC current converges to some limit value. Moreover, for $N \geq 800$, the results are affected only marginally by an increase of $N$ regardless of the particular choices of model parameters.

Before considering obtained results, let us briefly discuss the numerical solution of the system of equation (B.1) for cluster amplitudes. The set of nonlinear equations is solved by applying Newton’s iterative scheme. It is clear that the main computational cost comes from large buffer zones. Namely, for the cluster correlation superoperator quadratic with respect to
$b^\dagger$, $b^\ddagger$, the dimension of the system increases as $4N^2$. However, the system is sparse and the number of nonzero elements is proportional to $4N^2$. For $N = 800$, the system can be solved on a standard personal computer (e.g., 16 GB of RAM and Intel Core i5 quad-core CPU with 3.4 GHz clock speed) in about one hour. To solve the system, we have used sparse solver routines from Intel MKL FORTRAN library.

To illustrate the approach, we compute the current through the central region as a function of the level energy $\varepsilon_0$. In figure 2, each panel displays the current $J_L$ calculated for a particular choice of model parameters. We compare, for the same sets of model parameters, the calculated NECC(1) currents with those obtained by the second-order perturbation theory [8] and with the nonequilibrium configuration interaction method built on the coherent reference state (a so-called NECI* approximation) [11].

In the upper panels of figure 2, we consider the case where $N_a = 0$, which means the vibrational state in the system is not thermally excited. In this case, the NECC(1) current reaches a maximum value exactly at $\varepsilon_0 = \frac{\kappa^2}{\omega_0}$. It is the so-called reorganization energy\textsuperscript{4} of an electronic state associated with the electron–phonon coupling. Therefore, when $\varepsilon_0 = \varepsilon_0 - \frac{\kappa^2}{\omega_0}$ is close to chemical potentials of the electrodes (resonant regime), the NECC(1) current reaches its maximum value.

From the upper-left panel we see that for a weak electron–phonon coupling ($\kappa = 0.5$), all three approaches predict the currents that closely follow each other for all values of $\varepsilon_0$. This resemblance vanishes, however, with an increase in the coupling strength (see the upper-middle panel). In the resonant regimes, the SOPT and NECI* currents overestimate the

\textsuperscript{4} This reorganization energy is also referred to as polaron-shift [12, 13].
NECC result and, in addition, the SOPT current becomes an unphysical negative in the off-resonant regime. The negative SOPT current disappears if we consider the case of larger phonon energy (see the upper-right panel). For this case, the NECC and NECI* calculations give the same result, while the maximum value of the SOPT current noticeably exceeds the maximum values of the NECC(1) and NECI* currents.

The advantages of the NECC method become more apparent by considering the case where the central region contains a nonzero number of thermally excited equilibrium vibrational quanta. For $N_\omega = 1.0$, the results of such calculations are shown in the lower panels of figure 2. As is obvious from the graphs, the NECC(1) current is symmetric with respect to $\epsilon_0 = \kappa^2/\omega_0$. For the identical electrodes employed, this result is a consequence of a particle-hole symmetry of the whole system. Indeed, by applying the particle-hole transformation to all fermionic operators and then performing a shift of the phonon field, we get the same system but with an electronic state with energy $\epsilon_0' = -\epsilon_0 + 2\kappa^2/\omega_0$. Therefore, the current should be symmetric with respect to $\epsilon_0 = \kappa^2/\omega_0$.

Contrary to the NECC method, the SOPT and NECI* approaches do not fulfill the symmetry property. This is most clearly evident in the lower-middle panel of figure 2, where both the SOPT and NECI* approaches demonstrate the substantially asymmetric behaviour. It is interesting to note that the behaviour of the NECI* current becomes more symmetric when we decrease the coupling strength or increase the phonon energy. As a result, for $\kappa = 0.5$ the difference between the NECC(1) and NECI* currents is marginally negligible, while for $\kappa = 1.0$ and $\omega_0 = 2.0$ both approaches give results close to each other.

In figure 3 we compare NECC(1) and NECC(2) electron currents. We also show the current for the noninteracting ($\kappa = 0$) electronic level for the comparison in this figure. One can see from the figure that the inclusion of the higher-order term $S'$ [see equation (34)] into the cluster correlation superoperator does not substantially change the results of the NECC(1) approach. The difference between the NECC(1) and NECC(2) currents is negligible for weak coupling (left panel). Moreover, the inclusion of a four-fermion component into a cluster correlation superoperator leaves the $\epsilon_0$ dependence of the current almost unchanged even in the strong coupling regime (middle and right panels)—the difference between NECC(1) and NECC(2) is only a few percent of the total current. Thus, we can say that even the use of an excited cluster superoperator $S_1$, which is quadratic in the fermion-creation superoperators (33), captures the most of nonequilibrium correlations in our model.

5. Conclusions

In this paper, we propose an extension of the coupled cluster method to nonequilibrium correlated many-body quantum systems described by the Lindblad master equations. The method is based on a superoperator representation of the master equation. It was shown that in Liouville–Fock space the steady-state density matrix can be defined via the exponential ansatz $|\rho\rangle = \exp (S)|\rho_0\rangle$, where $|\rho_0\rangle$ is a reference state and $S$ is a cluster correlation superoperator linearly expanded in terms of a complete set of multiconfigurational creation superoperators. A general prescription of to calculate expansion (cluster) amplitudes was given.

As an application of the method, quantum transport through a vibrational electronic level was considered. To compute the steady-state current, a truncated expansion of the cluster correlation superoperator was used. It was shown that the method always preserves the continuity equation, no matter how we truncate the cluster correlation superoperator. It was demonstrated that the NECC method provides advantages over the perturbation theory and
the nonequilibrium configuration interaction method when the electron–phonon coupling is large or when the system contains thermally excited vibrational quanta.

To conclude we would like to mention some possible further steps. An obvious application of the NECC method is to apply it to the charge transport problem through the system with electron–electron correlations. Another route is to extend the method to a dynamical nonequilibrium case by making the cluster amplitudes time-dependent functions. All these will be the subjects of our future investigations.

Appendix A. Uncorrelated density matrix as a vacuum state

Here, we demonstrate how to define normal modes for the Liouvillian $L_0 = L_{el} + L_{ph} + L_B$, where

$$L_{el} = \varepsilon_0 \left( \hat{a}^{\dagger} \hat{a} - \hat{a} \hat{a}^{\dagger} \right), \quad L_{ph} = \omega_0 \left( \hat{d}^{\dagger} \hat{d} - \hat{d} \hat{d}^{\dagger} \right),$$

$$L_B = \sum_{\alpha \alpha \alpha} \varepsilon_{\alpha \alpha} \left( \hat{a}_{\alpha \alpha}^{\dagger} \hat{a}_{\alpha \alpha} - \hat{a}_{\alpha \alpha} \hat{a}_{\alpha \alpha}^{\dagger} \right) - i\Pi_{\alpha \alpha},$$

$$\Pi_{\alpha \alpha} = (\Gamma_{\alpha \alpha 1} - \Gamma_{\alpha \alpha 2}) \left( \hat{a}_{\alpha \alpha}^{\dagger} \hat{a}_{\alpha \alpha} + \hat{a}_{\alpha \alpha} \hat{a}_{\alpha \alpha}^{\dagger} \right) - 2i(\Gamma_{\alpha \alpha 1} \hat{a}_{\alpha \alpha}^{\dagger} \hat{a}_{\alpha \alpha} + \Gamma_{\alpha \alpha 2} \hat{a}_{\alpha \alpha} \hat{a}_{\alpha \alpha}^{\dagger}) + 2\Gamma_{\alpha \alpha 2}$$

such that the uncorrelated density matrix $|\rho_0\rangle = |\rho\rangle |\rho\rangle_B$ and the super-vector $\langle I |$ would be the right and left vacuum states, respectively.

First, we note that due to equation (4) the superoperators

$$\hat{\beta}^{\dagger} = \hat{a}^{\dagger} - i\tilde{a}, \quad \hat{\varphi}^{\dagger} = \hat{d}^{\dagger} - \hat{d}, \quad \hat{b}_{\alpha \alpha}^{\dagger} = \hat{a}_{\alpha \alpha}^{\dagger} - i\tilde{a}_{\alpha \alpha}$$

and their tilde-conjugated partners $\tilde{\beta}^{\dagger}$, $\tilde{\varphi}^{\dagger}$, $\tilde{b}_{\alpha \alpha}^{\dagger}$ annihilate $\langle I |$ when acting from the right. However, since $(\langle I |$ and $|\rho_0\rangle$) are not mutually Hermitian conjugates, the respective annihilation superoperators cannot be defined by taking the Hermitian conjugate of (A.2).
To define annihilation superoperators associated with the central region, we assume that they do not contain electrons when disconnected from the electrodes, while there are a certain number $N_\omega$ of thermally excited equilibrium vibrational quanta

$$\langle \hat{a}^\dagger \hat{a} | \rho \rangle_{el} = 0, \quad \langle \hat{a}^\dagger \hat{a} | \rho \rangle_{ph} = N_\omega. \quad (A.3)$$

Then, we introduce

$$\hat{\beta} = \hat{a}, \quad \hat{\beta} = \left( \hat{\beta} \right)^\dagger, \quad \hat{\gamma} = \left( 1 + N_\omega \right) \hat{d} - N_\omega \hat{d}^\dagger, \quad \hat{\gamma} = \left( \hat{\gamma} \right)^\dagger \quad (A.4)$$

such that $\hat{\beta}(\rho)_{el} = \hat{\beta}(\rho)_{cl} = 0, \hat{\gamma}(\rho)_{ph} = \hat{\gamma}(\rho)_{ph} = 0$ and

$$L_{el} = \epsilon_0 \left( \hat{\beta}^\dagger \hat{\beta} - \hat{\beta}^\dagger \hat{\beta} \right), \quad L_{ph} = \omega_0 \left( \hat{\gamma}^\dagger \hat{\gamma} - \hat{\gamma}^\dagger \hat{\gamma} \right). \quad (A.5)$$

As for the buffers, we can apply the equation of motion method and define

$$\hat{b}_{ka} = \left( 1 - f_{ka} \right) \hat{a}_{ka} - i f_{ka} \hat{a}_{ka}^\dagger, \quad \hat{b}_{ka} = \left( \hat{b}_{ka} \right)^\dagger \quad (A.6)$$

such that

$$L_B = \sum_{ka} \left( E_{ka} \hat{b}_{ka}^\dagger \hat{b}_{ka} - E_{ka} \hat{b}_{ka}^\dagger \hat{b}_{ka} \right). \quad (A.7)$$

where $E_{ka} = \epsilon_{ka} - i f_{ka}$. Thus, we have $\hat{b}_{ka}(\rho)_{B} = \hat{b}_{ka}(\rho)_{B} = 0$.

It should be recognized that although the above defined uncorrelated creation and annihilation operators are not Hermitian conjugates to each other, they satisfy the canonical (anti)commutation relations.

**Appendix B. System of nonlinear equations for coupled cluster amplitudes**

The system of nonlinear equations for the amplitudes in the cluster correlation operator $S_2$ has the following form

$$\sum_k f_k \left( I_k - I_k^* \right) = 0; \quad W q_0 + \kappa n = 0; \quad \eta_{10} q_0 - \sum_k f_k \left( I_{k10} - I_{k01}^* \right) + \kappa n (1 - n) = 0; \quad \eta_{k} \left( \epsilon_0 - E_k^* + 2 \kappa W \right) - t_k n - \sum_l f_{lk} F_{lk} + \kappa (I_{k10} + I_{k01}) = -t_k f_k;$$

$$I_{k10} \left( \epsilon_0 + \omega_0 - E_k^* + 2 \kappa W \right) - t_k n_{10} + \kappa I_k (1 - n + N_\omega) - \sum_l f_{lk10} = 0;$$
To obtain equations for the cluster amplitudes in $S_1$, we should neglect the last two equations and put $G_{i0} = 0$.

In the above equations, the nonequilibrium boundary conditions are introduced via fermionic occupation numbers $f_k$ that depend on the chemical potentials in the left ($k \in L$) and right electrodes ($k \in R$). Neglecting the electron–phonon correlations ($\kappa = 0$), we get the system of linear equations for the cluster amplitudes $n$, $I_k$, $F_{i0}$, which determine the exact steady-state density matrix for the charge transport problem through a noninteracting level.

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