NONEQUILIBRIUM TRANSPORT IN OPEN QUANTUM SYSTEMS*

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We propose a generalization of the nonequilibrium Green's functions to treatment of open systems, to account for the influence of the system-environment coupling on transport in nonequilibrium mesoscopic systems. Our approach is based on the partial-trace-free time-convolutionless equation of motion for the open system's reduced density matrix. We generalize the two-time correlation functions, and analyze the behavior of systems in the transient regime, as well as in a far-from-equilibrium steady state.

1. Introduction

In recent years, advances in mesoscopic physics and the miniaturization of semiconductor devices have increased the interest in theoretical treatment of electronic transport in small inhomogeneous structures[1,2]. Approaches based on the nonequilibrium Green's functions (NEGF), that assume open boundaries, have been successfully utilized in treatment of such devices[2]. However, the description of far-from-equilibrium situations in devices, in which a state achieved cannot be related uniquely to the initial state, poses some principal constraints on the application of the NEGF formalism[3,4]. Namely, it may be impossible to relate the far-from-equilibrium expectation values with those for the initial state, which may be, e.g., thermal equilibrium.[3] Also, there is the question of the validity of Wick's theorem in a given far-from-equilibrium state, which leads to difficulties with the diagrammatic expansion.[5]

A far-from-equilibrium steady state is achieved through a balance of the driving and the relaxation forces, and the driven system should be considered an open subsystem of a larger, interacting closed system. Unlike closed systems, for which the NEGF formalism is formulated, open systems typically obey non-Hamiltonian dynamics. Full treatment of open systems traditionally relies on the calculation of the system reduced density matrix, which is feasible only in fairly small systems, such as molecules. In many-body systems, the reduced density

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matrix is not only difficult to calculate, but actually contains too much information. Namely, for closed systems, it is known that the first few lowest-order Green's functions are usually quite sufficient to analyze electronic transport[1].

In this paper, we wish to generalize the Green's functions formalism to treatment of open many-body systems. Substantial work a few decades ago dealt with evaluations of multi-time correlation functions for open systems, but relied upon using the full system+environment evolution operators[6]. In order to avoid this feature, we utilize the partial-trace-free (PTF) time-convolutionless (TC) approach[7], based on a modification of the widely used projection operator technique[8], which allows us to properly define the open system's two-time correlation functions, without resorting to solving the full system+environment problem. We analyze the properties of transients, as well as far-from-equilibrium steady state conditions.

2. Two-Time Correlation Functions for Open Systems

We will be interested in generalizing the closed system's two-time correlation functions of the type $\mathrm{Tr}[\rho_H a_H(t') b_H(t)]$, with subscript H denoting the Heisenberg picture, to the case of an open system, whose dynamics is generally non-Hamiltonian. This will enable us to generalize the 'less-than' and 'greater-than' single particle Green's functions

$$iG^{<}(1,1') = \pm \mathrm{Tr}\left[\rho_H \psi_H^{+}(t') \psi_H(t)\right], \; iG^{>}(1,1') = \mathrm{Tr}\left[\rho_H \psi_H(t) \psi_H^{+}(t')\right], \qquad (1)$$

(upper sign refers to bosons, lower to fermions). The following approach can easily be generalized to higher-order Green's functions.

For a closed system with Hamiltonian $h(t)$, which may be time-dependent due to external driving forces, the density matrix ρ in the Schrödinger picture obeys the quantum Liouville equation

$$\frac{d\rho(t)}{dt} = -i\left[\, h(t), \rho(t)\,\right] \equiv -i\, L(t)\rho(t)\,, \qquad (2)$$

with L being the Liouville superoperator (superoperators will be denoted by capital letters). With T^{c} (T^{a}) denoting the chronological (anti-chronological) time-ordering operators and Θ being the Heaviside step-function, the solution of (2) is given by

$$\rho(t) = U(t,0)\rho(0)\,,$$

$$U(t,t') = \Theta(t-t')\, \mathrm{T}^{c}\, exp\left(-i\int_{t'}^{t} d\tau\, L(\tau)\right) + \Theta(t'-t)\, \mathrm{T}^{a}\, exp\left(i\int_{t}^{t'} d\tau\, L(\tau)\right). \qquad (3)$$

If a and b are time-independent operators in the Schrödinger picture, it is easily shown that the two-time correlation functions can be written as

$$\mathrm{Tr}\left[\rho_H\, a_H(t') b_H(t)\right] = \mathrm{Tr}\left\{ a\, U(t',t)\left[b\, \rho(t)\right]\right\}. \qquad (4)$$

Namely, when written in the Schrödinger picture, the desired expectation value actually means that, at time t, b acts on ρ (actually, b acts on the 'ket' part of ρ), then $b\rho(t)$ evolves under the quantum Liouville equation, until a acts on the result at time t'.

Now, consider an open system S, interacting with its environment E, so that the 'system+environment' (S+E) is closed, and possibly influenced by external driving fields that are assumed known and unaffected by the feedback from S+E. The Hilbert spaces of the environment and the system, $\mathcal{H}_E$ and $\mathcal{H}_S$, are assumed to be of finite dimensions d_E and d_S, respectively. The evolution of the total S+E density matrix ρ is given by the quantum Liouville equation (2), with the Hamiltonian h consisting of the system part $1_E \otimes h_S$, the environment part $h_E \otimes 1_S$ and the interaction part h_{int}, so that $h = 1_E \otimes h_S + h_E \otimes 1_S + h_{\mathrm{int}}$. Obviously, the corresponding Liouville operator is of the form $L = L_S + L_E + L_{int}$. The evolution of the system S is described by the reduced density matrix $\rho_S = Tr_E(\rho)$, with $Tr_E(...)$ denoting the partial trace over the environment states. To deduce how ρ_S evolves, we use the projection-operator technique[8], based on the uniform environment density matrix $\overline{\rho}_E \equiv d_E^{-1} \cdot 1_{d_E \times d_E}$, which introduces projection operators $\overline{P}$ and $\overline{Q}$ on $(\mathcal{H}_E \otimes \mathcal{H}_S)^2$ (the space of operators on the S+E Hilbert space $\mathcal{H}_E \otimes \mathcal{H}_S$) by the relations

$$\overline{P}x = \overline{\rho}_E \otimes \mathrm{Tr}_E x\,, \quad \overline{Q} = 1 - \overline{P}\,, \quad x \in (\mathcal{H}_E \otimes \mathcal{H}_S)^2\,. \qquad (5)$$

The above choice of the projection operators has a particular use. Namely, for a given basis $\{|\alpha\,\beta\rangle|\ \alpha, \beta = 1,...,d_S\}$ in $\mathcal{H}_S^2$, there is a simply constructed basis[7] $\{|\overline{\alpha\,\beta}\rangle|\ \alpha, \beta = 1,...,d_S\}$ in the unit eigenspace of $\overline{P}$, such that

$$\rho_S^{\alpha\beta} = (\mathrm{Tr}_E\, \rho)^{\alpha\beta} = \sqrt{d_E} \cdot (\overline{P}\,\rho)^{\overline{\alpha\beta}}\,. \qquad (6)$$

This feature enables us to track the evolution of ρ_S by writing the equation of motion for $\overline{P}\rho$ in the $\overline{P}$ eigenbasis, whose first d_S^2 vectors are $\{|\overline{\alpha\,\beta}\rangle\}$. This way, we avoid the standard procedure of taking the environment partial trace of

$\overline{P}\rho$, which means that we need not deal with full S+E evolution operators. By using the block forms in the eigenbasis of $\overline{P}$, we obtain

$$\overline{P}=\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix},\ \overline{Q}=\begin{bmatrix}0 & 0\\ 0 & 1\end{bmatrix},\ \rho=\begin{bmatrix}\rho_1\\ \rho_2\end{bmatrix},\quad \text{where}\ \rho_S=\sqrt{d_E}\,\rho_1 \tag{7}$$

and

$$L(t)=\begin{bmatrix}L_{11}(t) & L_{12}(t)\\ L_{21}(t) & L_{22}(t)\end{bmatrix},\ U(t,t')=\begin{bmatrix}U_{11}(t,t') & U_{12}(t,t')\\ U_{21}(t,t') & U_{22}(t,t')\end{bmatrix},\ \rho(t)=U(t,0)\rho(0). \tag{8}$$

This representation has several important features. First, one straightforwardly obtains that L_{11} is of the commutator-generated form, i.e. that it corresponds to an effective system Hamiltonian $h_{S,\mathrm{eff}}$, given by $h_{S,\mathrm{eff}}=h_S+\mathrm{Tr}_E(h_{int})/d_E$,[9] which accounts for the well-known first order correction to the system spectrum due to the coupling with the environment.[10] The terms that effectively account for the system-environment coupling are L_{12} and $L_{21}=(L_{12})^+$. In order for the system to evolve as decoupled from the environment, it must hold $\|L_{12}\rho_2\|<<\|L_{11}\rho_1\|$. This requirement is obviously fulfilled if the interaction vanishes (h_{int}=0 implies L_{12}=0). In Section 4, however, we will see that the above inequality can also be satisfied in the case of appreciable system-environment coupling, in a far-from-equilibrium steady state.

Multiplication of the total density matrix ρ by the system annihilation operator ψ_S can be described by the action of a superoperator $\Psi=I_E\otimes\Psi_S$ on ρ . However, any system superoperator (i.e., that of the form $I_E\otimes A_S$) commutes with $\overline{P}$, and it has a block-diagonal form in the eigenbasis of $\overline{P}$, such that its first block-matrix is A_S,[9] so we obtain

$$\Psi=\begin{bmatrix}\Psi_S & 0\\ 0 & \Psi_2\end{bmatrix}. \tag{9}$$

(The creation operator ψ_S^+ will be associated with Ψ^+). According to the discussion following (4), we see that, in order to define $G^<(1,1')$ properly, first Ψ acts on ρ at time t, and then $\Psi\rho$ evolves until t', when Ψ^+ will act on it. In particular, *we need only the system's view of this action*. Therefore, we define a quantity $\rho^{\psi,t}(t')$, corresponding to $\Psi\rho$ at t

$$\rho^{\psi,t}(t)=\Psi\rho(t)\ ,\ \rho^{\psi,t}(t')=U(t',t)\rho^{\psi,t}(t)\,. \tag{10}$$

Now we define

$$\begin{aligned}\pm iG_S^{<}(1,1') &\equiv \mathrm{Tr}_S\left[\Psi_S^{+}\rho_S^{\psi,t}(t')\right]\\ &= \mathrm{Tr}_S\left[\Psi_S^{+}U_{11}(t',t)\Psi_S^{+}\rho_S(t)\right]+\sqrt{d_E}\,\mathrm{Tr}_S\left[\Psi_S^{+}U_{12}(t',t)\Psi_2\,\rho_2(t)\right].\end{aligned} \tag{11}$$

The two terms on the right-hand-side (RHS) of (11) mean that there are two contributions to the given expectation value: one from the "purely system states" (the first term on the RHS, which reduces to (1) when the system-environment interaction is turned off (h_{int}=0)), and also a contribution from the "entangled' portion of the system states (the second term on the RHS). The second term is neglected when the system is approximated as closed, i.e. when the environment is assumed unaffected by the feedback from the system.[11]

In addition, by substituting $\Psi \to \Psi^{+}$ in (10) and (11), we obtain $\rho^{\psi^{+},t'}(t)$, and define

$$\begin{aligned}iG_S^{>}(1,1') &\equiv \mathrm{Tr}_S\left[\Psi_S\,\rho_S^{\psi^{+},t'}(t)\right]\\ &= \mathrm{Tr}_S\left[\Psi_S\,U_{11}(t,t')\Psi_S^{+}\rho_S(t')\right]+\sqrt{d_E}\,\mathrm{Tr}_S\left[\Psi_S\,U_{12}(t,t')\Psi_2^{+}\rho_2(t')\right].\end{aligned} \tag{12}$$

3. Transients

Evaluation of the evolution submatrices U_{ij} becomes difficult with increasing size of the system and environment, so a direct calculation is generally out of the question in real-life applications. However, within the time-convolutionless approach, U_{21} and U_{22} can be always written in terms of U_{11} and U_{12}. However, for this approach it is important to fix a point in time t_0, which enables the abovementioned relationships among U_{ij} to be formalized. In the analysis of transients, i.e., the processes during which the state of S+E can be tracked back to the initial density matrix $\rho(t_0)$, the time-convolutionless equations of motion for ρ_1 and ρ_2 are given by

$$\begin{aligned}\frac{d\rho_1(t)}{dt} &= -i\left[L_{11}(t)-L_{12}(t)\,K_{22}^{-1}(t;t_0)K_{21}(t;t_0)\right]\rho_1(t)\\ &\quad + i\,L_{12}(t)\,K_{22}^{-1}(t;t_0)H_{22}(t,t_0)\rho_2(t_0)\\ \rho_2(t) &= -K_{22}^{-1}(t;t_0)K_{21}(t;t_0)\rho_1(t)-K_{22}^{-1}(t;t_0)H_{22}(t,t_0)\rho_2(t_0)\,,\end{aligned} \tag{13}$$

where operators H_{22}, K_{21} and K_{22} satisfy

$$H_{22}(t,t') = T^{c}\,exp\left(-i\int_{t'}^{t} d\tau\, L_{22}(\tau)\right),$$

$$\frac{dK_{21}(t;t_0)}{dt} = -iL_{22}(t)K_{21}(t;t_0) + iK_{21}(t;t_0)L_{11}(t) + iK_{22}(t;t_0)L_{21}(t) \ , \qquad (14)$$
$$\frac{dK_{22}(t;t_0)}{dt} = -iL_{22}(t)K_{22}(t;t_0) + iK_{22}(t;t_0)L_{22}(t) + iK_{21}(t;t_0)L_{12}(t) \ ,$$
$$K_{21}(t_0;t_0) = 0 \ , \quad K_{22}(t_0;t_0) = 1 .$$

The equations of motion for the evolution submatrices *for a fixed t_0* are given by

$$\begin{aligned}
\frac{dU_{11}(t,t_0)}{dt} &= -i\left[L_{11}(t) - L_{12}(t)\ K_{22}^{-1}(t;t_0)K_{21}(t;t_0)\right]U_{11}(t,t_0) \\
\frac{dU_{12}(t,t_0)}{dt} &= -i\left[L_{11}(t) - L_{12}(t)\ K_{22}^{-1}(t;t_0)K_{21}(t;t_0)\right]U_{12}(t,0) \\
&\quad + iL_{12}(t)K_{22}^{-1}(t;t_0)H_{22}(t,t_0) \\
U_{21}(t,t_0) &= -K_{22}^{-1}(t;t_0)K_{21}(t;t_0)U_{11}(t,t_0), \\
U_{22}(t,t_0) &= K_{22}^{-1}(t;t_0)\left[H_{22}(t,t_0) - K_{21}(t;t_0)U_{12}(t,t_0)\right] .
\end{aligned} \qquad (15)$$

Now we are able to construct the two-time Green's functions, after solving (14) and (15), and taking into account $U(t',t) = U(t',t_0)U^+(t,t_0)$,

$$\begin{aligned}
\pm iG_S^<(1,1') &= \mathrm{Tr}_S\left[\Psi_S^+U_{11}(t',t)\Psi_S U_{11}(t,t_0)\rho_S(t_0)\right] \\
&\quad + \mathrm{Tr}_S\left[\Psi_S^+U_{12}(t',t)\Psi_2 U_{21}(t,t_0)\rho_S(t_0)\right] \\
&\quad + \sqrt{d_E}\,\mathrm{Tr}_S\left[\Psi_S^+U_{11}(t',t)\Psi_S U_{12}(t,t_0)\rho_2(t_0)\right] \\
&\quad + \sqrt{d_E}\,\mathrm{Tr}_S\left[\Psi_S^+U_{12}(t',t)\Psi_2 U_{22}(t,t_0)\rho_2(t_0)\right] .
\end{aligned} \qquad (16)$$

Equations (13)-(16) are undoubtedly complicated. Their main advantage, however, is enhanced transparency. There is no explicit environmental partial trace that obscures the structure of relevant terms, and an expansion in terms of L_{12} is straightforwardly performed. Furthermore, at very high temperatures, $\rho_2(t_0)$ may sometimes be approximated as zero (all weights in the environment density matrix become approximately equal, when for all the (relevant) environment energy levels $\varepsilon_{n,E}$, $\varepsilon_{n,E}/k_BT \to 0$), which eliminates the last two terms in $G^<$. We see that the equation of motion for ρ_S is then formally decoupled from the equation for ρ_2 , but the memory effects, accounted for through K_{21}, K_{22} in (13), are still present in the behavior. So, the memory effects are specified by the dynamics (i.e. coupling strength), and cannot be completely eliminated by a fortunate choice of an initial state, contrary to statements made in some works. As to how a diagrammatic expansion may be obtained, to account for the occurrence of decoherence in the transient regime, remains to be investigated.[9]

4. Far-from-Equilibrium Steady State

Reaching a well-controlled steady state, independent of initial conditions, is the goal of external driving forces in various systems, such as semiconductor devices. Typically, there exists a characteristic relaxation time τ_{relax}, after which a far-from-equilibrium steady state is achieved, i.e.,

$$\rho_S \approx const. \quad \left(t >> \tau_{\text{relax}}\right). \tag{17}$$

The relaxation time is sufficient to destroy the information about the initial correlations, and build up new ones, in agreement with the external driving forces. When the transients have died out, we assert that, since the relaxation forces have adjusted to the driving forces, no more information from the environment is being passed on to the system[9], i.e.,

$$\left\| L_{12}(t)\ \rho_2(t) \right\| << \left\| L_{11}(t)\ \rho_1 \right\| \quad (\, t >> \tau_{\text{relax}}\,). \tag{18}$$

The system effectively starts to be decoupled from the environment, and ρ_S evolves under L_{11} alone. Then, (17) actually means

$$\frac{d\rho_S}{dt} \approx -iL_{11}(t)\rho_S \approx 0 \quad \Leftrightarrow \left[h_{S,\text{eff}}(t), \rho_S\right] = \left[h_S(t) + \tfrac{1}{d_E}\text{Tr}_E h_{\text{int}}(t), \rho_S\right] \approx 0\,. \tag{19}$$

As a consequence of the decoupling requirement (18), the far-from-equilibrium steady-state $G^{<}$ takes on the typical form for closed systems, with the effective total system Hamiltonian $h_{S,\text{eff}}$, namely

$$\pm\, iG_S^{<}(1,1') = \text{Tr}_S\left[\Psi_S^{+} U_{11}(t',t)\Psi_S\, \rho_S\right] = \text{Tr}_S\left[\psi_S^{+}\, u(t',t)\psi_S \rho_S\, u(t,t')\right], \tag{20}$$

where

$$u(t',t) = \Theta(t'-t)\Gamma^{\text{c}}\, exp\left(-i\int_t^{t'} d\tau\, h_{S,\text{eff}}(\tau)\right) + \Theta(t-t')\Gamma^{\text{a}}\, exp\left(i\int_{t'}^{t} d\tau\, h_{S,\text{eff}}(\tau)\right). \tag{21}$$

Generalization of other single-particle Green's functions, as well as higher-order functions, are straightforward.

We are, in general, unable to solve (19). Furthermore, the solution may not be unique, and should further be specified by a relevant set of state parameters (such as occupation numbers, average energy, etc.). This can be accomplished by constructing an approximate *relevant statistical operator*[12], which satisfies the self-consistency requirements for the given set of state parameters. However, diagrammatic expansion in terms of the system-environment coupling $Tr_E(h_{int})/d_E$ may not be possible, as the relevant statistical operator does not, in general, admit Wick's decomposition. However, the Dyson equation can be recovered for the so-called *mixed Green's functions*.[13]

5. Conclusion

Presently, considerable effort is being focused on deepening our understanding of transport in various mesoscopic structures. In this paper, we introduce a generalization of the NEGF formalism for analysis of open systems, which aims at a more complete treatment of the relaxation mechanisms and decoherence in the essentially open mesoscopic systems. Based on the PTF equation of motion for the open system's reduced density matrix, the two-time correlation functions have been defined. The present approach offers a transparent perturbative treatment of the system-environment coupling, which ought to simplify the treatment of transients. In far-from-equilibrium situations, however, it is shown that the two-time correlation functions recover their closed-system forms, with an effective, modified system Hamiltonian.

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