Improved master equation approach to quantum transport: From Born to self-consistent Born approximation

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Beyond the second-order Born approximation, we develop an improved master equation approach to quantum transport by virtue of a self-consistent Born approximation. The basic idea is replacing the free Green’s function in the tunneling self-energy by an effective reduced propagator under the Born approximation. We found that the effect of this improvement is remarkable. For instance, completely beyond the scope of the conventional second-order master equation, the new approach can not only recover the exact result of noninteracting transport under arbitrary voltages, but also predict the challenging nonequilibrium Kondo effect. In addition to having an elegant structure, the application convenience and accuracy of the proposed scheme, as demonstrated by the examples in this work, suggest it a useful tool for quantum transports.

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I. INTRODUCTION

Landauer-Büttiker theory and the non-equilibrium Green’s function (nGF) approach are widely accepted as two typical formalisms for mesoscopic transports [1, 2]. Besides these two “orthodox” methods, we notice that the rate equation approach, an alternative choice, is particularly suited for transport though nanostructures with few discrete states. Straightforwardly, “classical” rate equation can be constructed by phenomenological considerations [3]. Later efforts include its derivation and quantum generalization in the context of the resonant tunneling system, based on the nGF quantum kinetic theory [4], as well as its modification to describe the quantum coherence broadly inhered in mesoscopic systems [4]. In particular, a microscopic derivation starting with the many-particle Schrödinger equation was also developed [4]. The obtained quantum rate equation can properly describe both the many-body Coulomb interaction and the quantum coherence inside the nano-system. However, the rate equation is valid only under large bias voltage.

For mesoscopic transport setup, if we regard the leads coupled to the central system as an environment, similiar to the consideration for a quantum open system, we can perform a perturbative expansion for the tunnel-coupling Hamiltonian, and obtain a master equation for the reduced density matrix (operator) after averaging the states of leads. Indeed, along this line, the perturbative master equation approach to quantum transport has been constructed [5, 6]. In most cases, such as the various quantum open systems as in quantum optics, we find that the perturbative master equation up to the 2nd-order expansion is good enough. In quantum transport, the 2nd-order expansion corresponds to, particularly, a sequential tunneling regime. Also, the 2nd-order perturbative master equation covers all the elements of the rate equation approach mentioned above. Moreover, the number-resolved version of the perturbative master equation [7], [8], has been demonstrated very successful for studying quantum noise and counting statistics in mesoscopic transport, including also the large-derivation analysis as proposed very recently [9].

Unfortunately, similar to the rate equation, the 2nd-order master equation does not include the tunneling-induced level-broadening effect, which implies then a validity condition only under large bias voltage. For transport through Coulomb-island system, on the other hand, while the 2nd-order master equation captures well the Coulomb staircase phenomenon, it cannot however describe the cotunneling process and the nonequilibrium Kondo effect. For the latter purposes, higher-order expansions of the tunneling Hamiltonian are needed, as presented in a variety of scenarios in literature [8, 10, 11, 12].

In the simpler case of transport through noninteracting systems, exact master-equation approach is possible [13]. In this approach, the non-perturbative Feynman-Vernon influence functional theory is employed to derive an exact master equation for the reduced density matrix, which is valid for general spectral density and for arbitrary temperatures and bias voltages. Using this exact master equation, non-Markovian transport behaviors can be investigated. For noninteracting transport under arbitrary bias voltage, other exact approaches, being equivalent to each other in essence, are also possible. For instance, the single electron wavefunction (SEWF) method [14], starting with a full Schrödinger equation, can evaluate ex-
actly the current contribution of an individual electron. The total current, in noninteracting cases, is thus an integration of the individual ones between the (arbitrary) voltage window.

Now we raise a question: is it possible to construct a simple master equation, being applicable to interacting system and to arbitrary voltages (not suffering from the restriction of large bias limit)? In this paper, we make an effort aiming to this goal. We notice that, actually, the 2nd-order master equation is obtained from the well-known Born approximation in the perturbative expansion of the tunneling Hamiltonian. The resultant dissipation term, in analogy to the quantum dissipative system, corresponds to a self-energy process of tunneling. On the other hand, it is well known that in Green’s function theory, an efficient correction to the self-energy diagram under Born approximation is by a self-consistent self-energy diagram, under the so-called self-consistent Born approximation (SCBA). Therefore, based on this insight, for the quantum transport we replace the free (system only) Green’s function in the 2nd-order self-energy diagram, with an effective propagator defined by the 2nd-order master equation, which favorably includes the tunneling self-energy in it. We found that the effect of this simple improvement is remarkable: it can recover not only the exact result of noninteracting transport under arbitrary voltages, but also the cotunneling and nonequilibrium Kondo effect in Coulomb interacting systems.

The paper is organized as follows. In Sec. II we present the main formulation of the master equation approach under SCBA, without specifying the concrete systems. The presentation in this central part constitutes a number of subsections: we first outline in II(A) the master equation approach under Born approximation to quantum transport, then discuss in II(B) the basic idea of the SCBA which is further implemented in II(C) to construct the improved master equation approach; subsequently, in II(D) and II(E), we consider the steady state and prove an exact equivalence with the nGFP approach for noninteracting systems. In Sec. III, we continue a challenging test for the proposed approach, by applying it to the transport through an interacting quantum dot, showing a recovery of the nonequilibrium Kondo effect. In Sec. IV we apply the approach to an Aharonov-Bohm double-dot interferometer, first recovering the exact result of noninteracting dots, then extending the study of interacting double-dots from large bias limit to arbitrary voltage. Finally, in Sec. V we summarize the work. As complementary materials, we arrange three Appendices (A, B and C) for some particular details which are omitted in the main text.

II. FORMULATION UNDER SCBA

A. Scheme under Born Approximation

Let us start with a transport setup described by

$$H = H_S(a_{\mu}^\dagger, a_\mu) + H_B + H'.$$  \hspace{1cm} (1)

In this Hamiltonian, $H_S$ is for the central system embedded between two leads which are regarded as a generalized bath (or reservoir) and are modeled by $H_B = \sum_{\alpha=L,R} \sum_{\mu} \epsilon_{\alpha \mu k} b_{\alpha \mu k}^\dagger b_{\alpha \mu k}$. And, the coupling between the system and the leads is described by the tunneling Hamiltonian, $H' = \sum_{\alpha=L,R} \sum_{\mu} (t_{\alpha \mu k} a_{\mu}^\dagger b_{\alpha \mu k} + H.c.)$. Here, $a_{\mu}^\dagger$ ($a_\mu$) is the electron creation (annihilation) operator of the system state “$\mu$”, while $b_{\alpha \mu k}^\dagger$ ($b_{\alpha \mu k}$) are the creation (annihilation) operators of the leads, associated with the labeled specific states.

Following the conventional treatment of quantum open systems, we introduce a collective reservoir (lead) operator coupled to the system state “$\mu$”, $f_{\alpha \mu} = \sum_k \epsilon_{\alpha \mu k} b_{\alpha \mu k}$, where $\alpha = L, R$. Using it, we can rewrite the tunneling Hamiltonian as $H' = \sum_{\alpha=L,R} \sum_{\mu} (a_{\mu}^\dagger f_{\alpha \mu} + H.c.)$. Then, treating $H'$ perturbatively up to the second-order, i.e., under the Born approximation, we formally obtain a master equation as

$$\dot{\rho}(t) = -i \mathcal{L} \rho(t) - \int_0^t d\tau \Sigma^{(2)}(t-\tau) \rho(\tau).$$   \hspace{1cm} (2)

Here, the reduced density matrix of the system, $\rho(t)$, is defined by tracing out the reservoir states from the entire system-plus-reservoir density matrix $\rho_{TR}(t)$, i.e., $\rho(t) = Tr_B[\rho_{TR}(t)]$. Two superoperators, the system Liouillian $\mathcal{L}$ and the 2nd-order self-energy superoperator $\Sigma^{(2)}$, are involved in Eq. (2), with explicit definitions as $\mathcal{L}(\cdots) = [H_S, \{ \cdots \}]$, and $\Sigma^{(2)}(t - \tau) = \langle \mathcal{L}'(t) \mathcal{G}(t, \tau) \mathcal{L}'(\tau) \rangle$. Here, $\mathcal{G}(t, \tau) = e^{-i \mathcal{L}(t-\tau)}$.

Reexpressing the Liouillian self-energy superoperator in a form in Hilbert-space, the integrand in Eq. (2) has four terms, see Appendix A and Fig. 1 where a diagrammatic technique on the real-time Keldysh contour is employed for an illustrative purpose. Or, in an algebraic form, we can obtain an explicit expression for the master equation as

$$\dot{\rho}(t) = -i \mathcal{L} \rho(t) - \sum_{\mu \sigma} \left\{ [a_{\mu}^\dagger, A_{\mu \sigma}(t)] + H.c. \right\}. \hspace{1cm} (3)$$

Here, for the sake of brevity, we make a couple of conventions: $\sigma = +$ and $-\sigma = -\sigma$; $a_\mu^\dagger = a_{\sigma \mu}^\dagger$ and $a_{\sigma \mu} = a_\mu$. Also, we denoted $A_{\mu \sigma}(t) = \sum_{\alpha=L,R} A^{(\sigma)}_{\alpha \mu \sigma}(t)$, while $A^{(\sigma)}_{\alpha \mu \rho}(t)$ reads

$$A^{(\sigma)}_{\alpha \mu \rho}(t) = \sum_{\nu} \int_0^t d\tau C^{(\sigma)}_{\alpha \mu \nu}(t - \tau) \{ \mathcal{G}(t, \tau) [a_\nu^\dagger \rho(\tau)] \},$$  \hspace{1cm} (4)
where the reservoir (lead) correlation functions are defined simply as $C_{\alpha\mu\nu}(t - \tau) = \langle f_{\alpha\mu}(t) f_{\alpha\nu}^\dagger(\tau) \rangle_B$. As introduced above, here we make similar convention that $f_{\alpha\mu}^{(+)}(t) = f_{\alpha\mu}(t)$ and $f_{\alpha\mu}^{(-)}(t) = f_{\alpha\mu}(t)$. The time dependence of these operators originates from using an interaction picture with respect to the Hamiltonian of leads, while the average $\langle \cdots \rangle_B$ is over the bath (leads) states.

It should be noted that, in deriving the above results, only the Born approximation was used, but not involving the Markovian approximation. The non-Markovian feature is manifest by the time non-local form in the self-energy terms, Eq. (4).

Finally, following Ref. [2], associated with the above master equation approach, the transport current simply reads
\[ I_\alpha(t) = \frac{2e}{\hbar} \sum_\mu \text{Re} \left\{ \text{Tr} \left[ A_{\alpha\mu\rho}^{(+)}(t) a_\mu - A_{\alpha\mu\rho}^{(-)}(t) a_\mu^\dagger \right] \right\}. \] (5)

Here, the trace is over the states of the central system.

**B. Basic Considerations**

In this subsection, we present the basic considerations that can lead us to an improved scheme of master equation. Strictly speaking, the 2nd-order master equation applies only to transport under large bias voltage. That is, the Fermi levels of the electrodes should be considerably further, at least several times of the level broadening $\Gamma$, away from the transport levels of the central system. This can be understood by the following simple example, say, resonant transport through a single-level quantum dot.

For this simple case, one can easily check that the 2nd-order master equation would predict a vanishing current at zero temperature, if the dot level $E_0$ is a little bit higher than $\mu_L (\mu_L > \mu_R)$. However, it is well known that a full quantum mechanical treatment will give a nonzero resonant tunneling current in this situation. Similar difficulty appears as well if the dot level $E_0$ is narrowly in between the voltage window (i.e., $\mu_L > E_0 > \mu_R$): no matter how small the Fermi levels are away from $E_0$, a full resonant current, $I = e\Gamma_L \Gamma_R / (\Gamma_L + \Gamma_R)$, will be predicted by the theory. Obviously, these unreasonable results are caused owing to the neglect of the level broadening effect.

We notice that the tunneling self-energy operator in Eq. (3), $\Sigma^{(2)}(t - \tau) = \langle L^\dagger(t) G(t, \tau) L^\dagger(\tau) \rangle$, contains a free (system only) Green’s function $G(t, \tau)$, which simply corresponds to, in the case of single-level dot, $e^{-iE_0(t - \tau)}$. Then, if we attach $e^{-i\Gamma(t - \tau)}$ to this unitary propagator, the level ($E_0$) broadening effect is restored. Insightfully, the inserted factor, $e^{-i\Gamma(t - \tau)}$, represents nothing but a self-consistent self-energy correction. That is, it improves the self-energy diagram, using a Feynman diagrammatic language, from the Born to a self-consistent Born approximation.

In Green’s function theory, the correction of the self-energy diagram under self-consistent Born approximation (SCBA) is typically an efficient technique. In Fig. 1(A), taking the electron-phonon (or photon) interaction as an example, we show the basic idea of such correction. There, the single (solid) line is for the free Green’s function $G_0$ of the electron, the double-lines for its full Green’s function $G$, and the dashed line for the Green’s function of the phonon (or photon). The basic idea of the self-consistent Born approximation is to replace $G_0$ in the self-energy diagram by an improved one, $G^{(2)}$, as schematically shown in Fig. 1(A). It is well known that this correction corresponds to a systematic re-summation of the perturbation and can greatly improve the results in most cases.

For the 2nd-order master equation, Eq. (3), the Keldysh diagrammatic representation of the tunneling self-energy term is shown in Fig. 1(B). Analogously, within the same spirit of the self-consistent Born approximation, an improved treatment for the tunneling perturbation is proposed in this work, by replacing the free (system only) propagator $G(t, \tau)$ with an effective one to include the tunneling self-energy in it, as schematically shown in Fig. 1(C). The resultant scheme, in the remaining parts of this work, is to be termed as SCBA master equation (SCBA-ME). In next subsection, we shall present the detailed formulation simply based on this type of observation.

**C. Improved Scheme under SCBA**

Based on Eq. (3) we formally introduce an evolution operator $U(t, \tau)$, which propagates the state in terms of $\rho(t) = U(t, \tau) \rho(\tau)$. Then, the insight gained above leads us to replacing $G(t, \tau)$ with $U(t, \tau)$, in the self-energy operator or more precisely in $A_{\alpha\mu\rho}^{(2)}$ [c.f. Eq. (3)]:
\[ A_{\alpha\mu\rho}^{(\sigma)}(t) = \sum_\nu \int_0^t d\tau C_{\alpha\nu\rho}(t - \tau) \{ U(t, \tau) [ a_\nu^\dagger \rho(\tau) \} \}. \] (6)

Incorporating this improved quantity into the master equation and the transport current, we have
\[ \dot{\rho}(t) = -i\mathcal{L}\rho(t) - \sum_{\mu\sigma} \left\{ [a_\mu^\dagger, A_{\alpha\rho}^{(\sigma)}(t)] + \text{H.c.} \right\}, \] (7)
and
\[ I_\alpha(t) = \frac{2e}{\hbar} \sum_\mu \text{Re} \left\{ \text{Tr} \left[ A_{\alpha\mu\rho}^{(+)}(t) a_\mu - A_{\alpha\mu\rho}^{(-)}(t) a_\mu^\dagger \right] \right\}. \] (8)

We notice that, desirably, Eqs. (3) and (7) have the same compact structures as their counterparts under the simple 2nd-order Born approximation. The only difference is the replacement of $A_{\alpha\mu\rho}^{(\pm)}(t)$ by $A_{\alpha\mu\rho}^{(\pm)}(t)$. The most obvious consequence of this replacement is that the broadening effect, induced by the tunneling processes, is
FIG. 1: (color online) (A): The self-consistent Born approximation in Green’s function theory, where the free Green’s function $G_0$ is replaced by an effective one, $G^{(2)}$. The dashed line represents, for instance, the Green’s function of phonon/photon for an electron-phonon/photon interacting system. (B): The 2nd-order tunneling self-energy diagram, $\Sigma^{(2)}(t-\tau)$, in the real-time Keldysh contour representation. The dashed line represents the reservoir electron Green’s function. (C): The improved tunneling self-energy diagram under the self-consistent Born approximation, in which the free (system only) Green’s function $G(t,\tau)$ was replaced by the 2nd-order effective propagator $U(t,\tau)$.

included in the system state evolution in the self-energy terms of the master equation. But, not only limited to this, it has more implications. For instance, a careful inspection of Fig. 1(C) reveals that this replacement, significantly, accounts for the interplay of the multiple tunneling processes and the Coulomb interactions inside the central system. It is well known that such type of interplay is the basic reason for Kondo effect, including particularly the nonequilibrium Kondo effect in quantum transport though Anderson-type impurities. In Sec. III we will detail an example for this understanding. Moreover, it can be proved that cotunneling processes are most naturally contained in the proposed SCBA-ME scheme. Under the bias condition of Coulomb blockade, the SCBA-ME can recover the cotunneling results given by other approaches\cite{11}.

Below we outline a protocol of solving Eq. (6), in frequency domain, by the use of Laplace transformation: $\rho(\omega) = L[\rho(t)] = \int_0^{\infty} e^{-i\omega t}\rho(t)dt$. Performing the Laplace transformation on Eq. (6), we obtain

\[
- i\omega \rho(\omega) - \rho(0) = -i\mathcal{L}_{\rho}(\omega) - \sum_{\mu\sigma} \left\{ [a_{\mu\sigma}^\dagger, A^{(\sigma)}_{\mu\sigma}(\omega)] - [a_{\mu\sigma}, A^{(\sigma)}_{\mu\sigma}(\omega)] \right\} ,
\]

where $A^{(\sigma)}_{\mu\rho}(\omega) = \sum_{\sigma'} A^{(\sigma)}_{\mu\rho}(\omega)$, with $A^{(\sigma)}_{\mu\rho}(\omega)$ explicitly expressed as

\[
A^{(\pm)}_{\alpha\mu\nu}(\omega) = \sum_\nu \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \Gamma^{(\pm)}_{\alpha\mu\nu}(\omega') U(\omega \pm \omega')[a_{\nu}^\dagger \rho(\omega)].
\]

In deriving this result, we used the simple relation $L[e^{\pm i\omega't}U(t)] = U(\omega \pm \omega')$, and the Fourier expansion $C^{(\pm)}_{\alpha\mu\nu}(t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{\pm i\omega t} \Gamma^{(\pm)}_{\alpha\mu\nu}(\omega)$. In the latter expression, we introduce $\Gamma^{(\pm)}_{\alpha\mu\nu}(\omega) = \Gamma^{(\pm)}_{\alpha\mu\nu}(\omega f^{(\pm)}(\omega))$ and $\Gamma^{(c)}_{\alpha\mu\nu}(\omega) = \Gamma^{(c)}_{\alpha\mu\nu}(\omega f^{(c)}(\omega))$, where $\Gamma^{(c)}_{\alpha\mu\nu}(\omega) = 2\pi \sum_k \epsilon_{\alpha\mu k} \epsilon_{\alpha\nu k} \delta(\omega - \epsilon_k) f^{(c)}(\omega) = f^{(c)}(\omega)$ and $f^{(c)}(\omega) = 1 - f^{(c)}(\omega)$ are, respectively, the spectral density function of leads and the occupied and unoccupied Fermi functions.

Eqs. (9) and (11) constitute a closed form of coupled equations, which allows for straightforward evaluation for the solution in frequency domain. However, in doing this, we must explicitly identify the propagator $U(\omega)$ in Eq. (11). More specifically, we need to consider the evolution of $\tilde{\rho}_j(t) \equiv U(t, t_0)\rho(t_0)$. Here, for brevity, we use a single index "j" to denote the double ones, "{\nu, \sigma}". In this context, a special care need to be emphasized. Originally, the 2nd-order reduced propagator $U$ was introduced (in time domain) from the usual propagation of a physical state, i.e., $\rho(t) = U(t, t_0)\rho(t_0)$. However, in Eq. (9) or (11), the quantity being propagated is $a_{\nu\sigma}^\dagger \rho$, which is not a physical state. Mathematically, we may expect from the well-known experiences that the propagator must be independent of the initial condition, which is, in the present context, the object (initial state) to be propagated. In most cases, this statement is true. However, the analysis in Appendix A shows that this "general rule", quite unexpectedly, breaks down in our case. In Appendix A, we prove that, the evolution rule (equation) of $\tilde{\rho}_j(t)$ differs from the one for $\rho(t)$. To our knowledge, this is the first time to recognize this subtle issue in constructing a master equation. We will see that, in the following illustrative examples, this clarification is extremely important – otherwise we cannot get the correct results.

Now we summarize the result derived in Appendix A as follows:

\[
\tilde{\rho}_j(t) = -i\mathcal{L}_{\tilde{\rho}}(t) - \sum_{\mu} \left\{ [a_{\mu}, A^{(\pm)}_{\mu\rho}] + [a_{\mu}^\dagger, A^{(c)}_{\mu\rho}] \right\} \]

\[
+ \left\{ [a_{\mu}^\dagger, A^{(\pm)}_{\mu\rho}] + [a_{\mu}, A^{(c)}_{\mu\rho}] \right\} .
\]

The operators $A^{(\pm)}_{\mu\rho}$ in this equation are actually the same as $A^{(\pm)}_{\nu\sigma}$ [c.f. Eq. (9)], only replacing $\rho$ by $\tilde{\rho}_j$. Out of our expectation, we notice a significant difference between Eq. (11) and Eq. (6): the commutators in the standard master equation (6) become now the anti-commutators in Eq. (11)!

The solution of Eq. (11) determines the propagator $U(\omega)$ in Eq. (11). Therefore, we Laplace transform
Eq. (11):

\[ -i\omega \hat{\rho}_j(\omega) - \hat{\rho}_j(0) = -i\mathcal{L}\hat{\rho}_j(\omega) - \Sigma(\omega)\hat{\rho}_j(\omega). \]  

Here, in frequency domain, the self-energy superoperator reads

\[ \Sigma(\omega) = \sum_{\sigma\mu} \left[ \tilde{a}_\sigma^\dagger C_{\sigma\mu}(\omega - \mathcal{L})a_\mu^\dagger + \tilde{a}_\sigma a_\mu C_{\sigma\mu}^*(\mathcal{L} - \omega)a_\mu^\dagger + a_\mu C_{\sigma\mu}(\omega - \mathcal{L})a_\mu^\dagger + a_\mu C_{\sigma\mu}^*(\mathcal{L} - \omega)a_\mu^\dagger \right]. \]  

In this equation, the shorthand notations \( \tilde{a}_\sigma^\dagger \hat{O} = a_\sigma^\dagger \hat{O} \) and \( a_\sigma^\dagger \hat{O} \equiv \hat{O}a_\sigma^\dagger \), considerably simplify the result. And, \( C_{\alpha\mu}(\omega), C_{\sigma\mu}^*(\omega) \), the Laplace transformation of \( C_{\alpha\mu}(t) \) are related with \( \Gamma_{\alpha\mu}(\omega) \) through the well known dispersive relation:

\[ C_{\alpha\mu}(\omega) = \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \frac{i}{\omega + \omega' + i\tau} \Gamma_{\alpha\mu}(\omega). \]  

Eqs. (6), (7) and (12) constitute the main formulation of the SCBA master equation. In Appendix B we prove a relation of the SCBA-ME with an alternative approach developed recently, say, the hierarchical master equations (HMEs) scheme \[13\]. The HMEs, in terms of a hierarchical equation-of-motion (HEOM) for the density operator and a series of auxiliary operators, is derived by a series of derivatives on the Feynman-Vernon influence functional. Since the SCBA-ME scheme largely depends on a spectral decomposition technique, this type of construction makes the numerical implementation very tricky and time-consuming. In Appendix B we show that the SCBA master equation contains, significantly, the dominant contribution (terms) of the HMEs. This identification indicates that the SCBA-ME scheme should be a promising tool for quantum transports. In addition to the advantage with transparent physics, the SCBA-ME approach also allows for devising more efficient computational schemes, by virtue of its compact and closed-form structure.

Based on Eqs. (9), (11) and (12), one can actually carry out the time-dependent solution. In practice, from Eq. (12), we first calculate and store the reduced evolution operator, \( \mathcal{U}(\omega_m) \), for a series of frequencies \( \omega_m \). Then, we solve the coupled Eqs. (9) and (11) with the help of \( \mathcal{U}(\omega_m) \) to evaluate the integral in Eq. (10). Using the solution (in frequency domain), an inverse Laplace transformation then gives the system state \( \rho(t) \) and the transient current \( I(t) \), simultaneously. In this context, we would like to mention that, instead of the Fourier expansion for \( C_{\alpha\mu}(t) \), we can employ the Padé decomposition technique as exploited in Ref. \[24\] to improve the algorithm efficiency. Since this technique can replace the integral \( \int_{-\infty}^{\infty} d\omega' \) in Eq. (10) by a discrete sum, favorably, it reduces the numbers of \( \mathcal{U}(\omega_m) \).

D. Steady State

Based on Eqs. (9), (11) and (12) the steady-state result can be more easily obtained than the above transient solution. For this purpose, let us first consider the integral in Eq. (11). Since physically, the time correlation function in the integrand of Eq. (11) is nonzero only on \textit{finite} timescale (in Markovian limit which is in fact very short), we can then replace \( \rho(\tau) \) in the integrand by the steady-state density matrix \( \bar{\rho} \), in the long time limit, \( t \rightarrow \infty \). After this replacement, we first make a Fourier expansion for \( C_{\alpha\mu}(t - \tau) \), then Laplace transform \( \mathcal{U}(t - \tau)[a_\sigma^\dagger \bar{\rho} \]}

\[ A_{\alpha\mu}(t \rightarrow \infty) = \sum_{\nu} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Gamma_{\alpha\mu}(\omega) \mathcal{U}(\pm\omega)[a_\nu^\dagger \bar{\rho}], \]  

Together with Eq. (13), substituting this result into Eq. (9), we can then straightforwardly solve for \( \bar{\rho} \) and calculate the steady-state current.

Now, in order to provide further insight for the proposed scheme, in the steady-state case, we recast the current formula Eq. (8) into a more conventional form. To this end, we introduce: \( \varphi_{1\mu}(\omega) = \text{tr}[a_\mu^\dagger \rho_{1\mu}(\omega)] \), and \( \varphi_{2\mu\nu}(\omega) = \text{tr}[a_\mu^\dagger \rho_{2\mu\nu}(\omega)] \), where \( \rho_{1\mu}(\omega) \) and \( \rho_{2\mu\nu}(\omega) \) are the solutions of Eq. (12), with an initial condition \( \rho_{1\mu}(0) = a_\mu^\dagger \bar{\rho} \) and \( \rho_{2\mu\nu}(0) = a_\mu^\dagger a_\nu \bar{\rho} \), respectively. Moreover, we introduce a matrix notation using, for instance, \( \varphi \) to denote the matrix with elements \( \varphi_{\mu\nu} \). Then, the steady-state current can be expressed in a compact form as

\[ I_\alpha = \frac{2e}{h} \text{Re} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \text{Tr}\{\Gamma_\alpha(\omega)[f_\alpha(\omega)\varphi(\omega) - \varphi_1(\omega)]\}, \]  

where \( \varphi(\omega) = \varphi_1(\omega) + \varphi_2(\omega) \).

Further simplification is possible, if \( \Gamma_L = \lambda \Gamma_R \), where \( \lambda \) is a constant. In this case, Eq. (14) can be recast to the Landauer-Büttiker type of current formula. That is, the current is an integration of tunneling coefficient over the bias window: \( I = \frac{2e}{h} \text{Re} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} [f_L(\omega) - f_R(\omega)] T(\omega) \). In our case, the effective tunneling coefficient reads \( T(\omega) = \text{Tr}\{\Gamma_L \Gamma_R (\Gamma_L + \Gamma_R)^{-1} \text{Re}[\varphi(\omega)]\} \). Compared to the result of nGF formulation \[4\], we find that \( \varphi \) plays a role of the retarded Green’s function, i.e., \( \varphi(\omega) = iG^R(\omega) \). The point is that, the current formula in terms of nGF is only a formal expression: it does not hint anything for the methods to obtain the various Green’s functions. Our \( \varphi \), however, is based on a concrete computational scheme in terms of a master equation approach. It is therefore of interest to investigate to what extent the proposed master equation scheme under SCBA is precise. In the follow-up, we will demonstrate: (i) it is exact for noninteracting systems, and (ii) it is \textit{good enough} for interacting systems – it can predict, for instance, the nonequilibrium Kondo effect.
E. Noninteracting System: Recovery of the Exact Result under Arbitrary Bias

Consider, in this subsection, the transport through a noninteracting multi-level system, \( H_S = \sum_{\mu \nu} h_{\mu \nu} a_{\mu}^{\dagger} a_{\nu} \). Straightforwardly, based on Eq. (12), we obtain the equation of motion for \( \varphi_i \), as follows:

\[
-i\omega \varphi_i(\omega) - \varphi_i(0) = -i\hbar \varphi_i(\omega) - i\Sigma_0(\omega)\varphi_i(\omega).
\]

(17)

\( \varphi_i(0) \) stand for the initial conditions, \( \varphi_{1\mu \nu}(0) = \text{Tr}[a_{\mu} \hat{\rho} a_{\nu}^{\dagger}] \) and \( \varphi_{2\mu \nu}(0) = \text{Tr}[a_{\mu} a_{\nu}^{\dagger} \hat{\rho}] \). The tunnel-coupling self-energy \( \Sigma_0 \) reads \( \Sigma_{0\mu \nu}(\omega) = -i \int d\omega' [C_{\alpha \mu \nu}^{(-)}(\omega) + C_{\alpha \mu \nu}^{(+)}(-\omega)] \), or

\[
\Sigma_{0\mu \nu}(\omega) = \int_{-\infty}^{\infty} d\omega' \frac{\Gamma_{\mu \nu}(\omega')}{2\pi}.
\]

(18)

Then, based on Eq. (17), summing up \( \varphi_1(\omega) \) and \( \varphi_2(\omega) \) yields

\[
\varphi(\omega) = i\left[\omega - \hbar - \Sigma_0(\omega)\right]^{-1} iG'(\omega).
\]

(19)

In deriving this result, the cyclic property under trace and the anti-commutator, \( \{a_{\mu}, a_{\nu}^{\dagger}\} = \delta_{\mu \nu} \), have been used.

Eq. (19) is the exact Green’s function of transport through a noninteracting system. We then conclude that our improved master equation approach under SCBA is exact for noninteracting transports. Inserting Eq. (19) into the steady-state current formula given above, we can evaluate the current for arbitrary bias voltage. Therefore, very importantly, this achievement is beyond the ability of the usual 2nd-order master equation approach, which applies only to large bias limit, even for the case of noninteracting transport.

III. TRANSPORT THROUGH AN INTERACTING QUANTUM DOT

In this section, we perform a challenging test for the master equation under SCBA. That is, we consider the transport through a strongly interacting quantum dot, modeled by the well-known Anderson impurity Hamiltonian:

\[
H_S = \sum_{\mu} \left( \epsilon_{\mu} a_{\mu}^{\dagger} a_{\mu} + \frac{U}{2} n_{\mu} n_{\bar{\mu}} \right).
\]

(20)

The index \( \mu \) labels the spin up (“↑”) and spin down (“↓”) states, and \( \bar{\mu} \) stands for the opposite spin orientation. \( \epsilon_{\mu} \) denotes the spin-dependent energy level, which may account for the Zeeman splitting in the presence of magnetic field \( (B) \), \( \epsilon_{\uparrow, \downarrow} = \epsilon_0 \pm g \mu_B B \). Here \( \epsilon_0 \) is the degenerate dot level in the absence of magnetic field; \( g \) and \( \mu_B \) are, respectively, the Lande-\( g \) factor and the Bohr’s magneton. In the interaction part, say, the Hubbard term \( U \eta n_{\uparrow} n_{\bar{\mu}} = a_{\mu}^{\dagger} a_{\mu} a_{\bar{\mu}}^{\dagger} a_{\bar{\mu}} \) is the number operator and \( U \) represents the interacting strength.

Applying the SCBA-ME approach to this system, we first note that the correlation function, \( C^{(\pm)}_{\alpha \mu \nu}(t) \), is diagonal with respect to the spin states, i.e., \( C^{(\pm)}_{\alpha \mu \nu}(t) = \delta_{\mu \nu} C^{(\pm)}_{\alpha \mu \nu}(t) \), and \( G^{(\pm)}_{\alpha \mu \nu} = G^{(\pm)}_{\alpha \mu \nu} \delta_{\mu \nu} \). Then, we specify the Hilbert space as follows. Simply, there are four states involved in the transport: \( |0\rangle \), \( |\uparrow\rangle \), \( |\downarrow\rangle \) and \( |d\rangle \), corresponding to, respectively, the empty, spin-up, spin-down and double occupancy states. Using this state basis, we can reexpress the electron operator in terms of a projection operator, \( a_{\mu}^{\dagger} = |\mu\rangle \langle 0| + (-1)^{\mu} |d\rangle \langle \bar{\mu}| \), where the conventions \((-1)^{\uparrow} = 1 \) and \((-1)^{\downarrow} = -1 \) are implied.

For a solution of steady state, as Eq. (15), we have

\[
J^{(\pm)}_{\alpha \mu \bar{\nu}} = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Gamma^{(\pm)}_{\alpha \mu \nu}(\omega) U(\pm \omega) a_{\mu}^{\dagger} a_{\nu}^{\dagger} a_{\mu}^{\dagger} a_{\nu} A_{\mu \nu \bar{\nu}}.\]

(21)

Straightforwardly, after some algebra, \( U(\pm \omega) a_{\mu}^{\dagger} a_{\nu}^{\dagger} a_{\mu}^{\dagger} a_{\nu} A_{\mu \nu \bar{\nu}} \) can be carried out as

\[
U(\omega) a_{\mu}^{\dagger} a_{\nu}^{\dagger} = \left[ \lambda_{\mu}^{\dagger}(\omega)|\mu\rangle \langle 0| + \kappa_{\mu}^{\dagger}(\omega)(-1)^{\mu} |d\rangle \langle \bar{\mu}| \right],
\]

\[
U(-\omega) a_{\mu}^{\dagger} a_{\nu}^{\dagger} = \left[ \lambda_{\mu}^{\dagger}(\omega)|\mu\rangle \langle 0| + \kappa_{\mu}^{\dagger}(\omega)(-1)^{\mu} |d\rangle \langle \bar{\mu}| \right].
\]

(22)
Then, we find the solution of \( \varphi(\omega) \) as
\[
\varphi(\omega) = \frac{i\Gamma_{\mu}(\omega) - \Sigma_{\mu}(\omega)}{\Pi_{\mu}(\omega) - \Sigma_{\mu}(\omega)}(1 - n_{\mu}) \\
\frac{i\Gamma_{\mu}(\omega) - \Sigma_{\mu}(\omega)}{\Pi_{\mu}(\omega) - \Sigma_{\mu}(\omega)} + \frac{i\Gamma_{\mu}(\omega) - \Sigma_{\mu}(\omega)}{\Pi_{\mu}(\omega) - \Sigma_{\mu}(\omega)} n_{\mu} \\
= \frac{\omega - \mu}{\Pi_{\mu}(\omega) - \Sigma_{\mu}(\omega)}(1 - n_{\mu}) \\
\frac{\omega - \mu}{\Pi_{\mu}(\omega) - \Sigma_{\mu}(\omega)} + \frac{\omega - \mu}{\Pi_{\mu}(\omega) - \Sigma_{\mu}(\omega)} n_{\mu}. \\
(24)
\]

In this result, \( n_{\mu} = \rho_{\mu} + \rho_{dd} \) and \( 1 - n_{\mu} = \rho_{\mu} + \rho_{dd} \). Eq. (24), precisely, coincides with the result by an equation-of-motion technique of nonequilibrium Green’s function [5].

The solution Eq. (24) contains the remarkable nonequilibrium Kondo effect, by the terms \( U\Sigma_{\mu}(\cdots)^{-1} \) in the denominators of \( \varphi(\omega) \). In Fig. 2 we demonstrate numerically the Kondo peaks in the differential conductance. At high temperatures, the terms \( U\Sigma_{\mu}(\cdots)^{-1} \) would vanish, reducing Eq. (24) to
\[
\varphi_{HF}(\omega) = \frac{i(1 - n_{\mu})}{\omega - \mu} + \frac{i\rho_{\mu}}{\omega - \mu - U - \Sigma_{0\mu}}. \\
(25)
\]

Here we use \( \varphi_{HF}(\omega) \), instead of \( \varphi(\omega) \), to imply the result being at the level of a mean-field Hatree-Fock approximation. As a matter of fact, Eq. (24) can be derived from the equation-of-motion technique of nGF at lower-order cutoff, by using a mean-field approximation [4]. However, even this simple result goes beyond the scope of the 2nd-order master equation, by noting that it contains the *broadening effect*. In Fig. 3 we plot the current-voltage relation based on Eq. (24) against that from Eq. (25). Here we emphasize that, in the Coulomb-blockade regime, i.e., in the plateau stages, the contribution of cotunneling processes has been automatically included in Eq. (24).

**IV. TRANSPORT THROUGH DOUBLE DOTS IN PARALLEL**

In this section we consider a more complex setup which consists of double (quantum) dots (DD) connected in parallel to the leads. Moreover, the interference loop is pierced by a magnetic flux (\( \Phi \)), similar to the well known Aharonov-Bohm interferometer. It was shown by the recent studies [22] that this system holds some interesting properties such as non-analytic current switching behavior, anomalous phase shift, and giant fluctuations of current. And, all of these are induced by an interplay of the inter-dot Coulomb correlation and quantum interference. In the present work we extend the study on this system from the large bias limit of Ref. [22], to arbitrary bias voltage.
Following Ref. [22], we assume that each dot has only one level, $E_{1(2)}$, involved in the transport. For simplicity, we also neglect the spin degrees of freedom, whose effect, under strong Coulomb interaction, can be easily restored by doubling the tunneling rates of each dot with the left lead [22]. Under these considerations, the DD Hamiltonian reads

$$H_{DD} = \sum_{\mu=1,2} E_{\mu} a_{\mu}^\dagger a_{\mu} + U a_{1}^\dagger a_{1} a_{2}^\dagger a_{2}. \quad (26)$$

Here $a_{1,2}^\dagger$ are the creation operators of the DD states, and the second term describes the interdot repulsive interaction. As in Eq. (1), we describe the leads by $H_L = \sum_k \epsilon_{L} b_k^\dagger b_k + \sum_{\mu} t_{L,\mu} a_{\mu}^\dagger b_k + t_{R,\mu} b_k^\dagger a_{\mu} + H.c.$.

Usually, the coupling amplitudes $t_{L(R),\mu}$ are treated being energy independent. However, in some circumstances certain difficulty such as divergence will arise from this assumption. To remedy this drawback, one may adopt an energy-dependent coupling amplitudes, as to be discussed in the following, in the interacting case for more details.

In the absence of magnetic field one can always choose the gauge in such a way that all couplings are real. In the presence of a magnetic flux $\Phi$, however, the tunneling amplitudes between the dots and the leads are in general complex. We write $t_{L(R),\mu} = \tilde{t}_{L(R),\mu} e^{i \phi_{L(R),\mu}}$, where $\tilde{t}_{L(R),\mu}$ is the coupling without the magnetic field. The phases are constrained to satisfy $\phi_{L1} + \phi_{R1} - \phi_{L2} - \phi_{R2} = \phi$, where $\phi \equiv 2\pi \Phi/\Phi_0$. In this work, we only consider the case with strong inter-dot Coulomb blockade($E_{1,2} + U \gg \mu_L$)(two dots cannot be both occupied simultaneously).

## A. Noninteracting Case

We would like to remind that, the usual 2nd-order master equation under Born approximation, is applicable only for large bias voltage. To demonstrate the applicability of our master equation under SCBA for arbitrary voltages, we first consider a noninteracting case, $U = 0$, which may correspond to the double dots being more separated away.

For this noninteracting system, in principle, a standard EOM technique in the framework of nonequilibrium Green’s function can carry out an exact solution [2]. Alternatively, in Ref. [22], using a full Schrödinger equation based single-electron-wave-function (SEWF) method, simple analytic result was obtained in a rather transparent way. The basic idea is that, in the absence of Coulomb correlation, the current is contributed independently by each individual electron from the source lead, with energy $\epsilon_{L,k}$ in the window from $\mu_R$ to $\mu_L$ (at zero temperature). That is, $I(t) = \int_{\mu_L}^{\mu_R} \rho_{L,k}(t) I_L^k(t)$, where $\rho_{L,k}$ is the density-of-states of the left lead. Moreover, the SEWF approach allows one to calculate $I_L^k(t)$ from a time dependent Schrödinger equation for the entire system (dots plus leads). Taking $\tilde{t}_{L(R)} = \tilde{t}_{L(R)}^{\mu} = \tilde{t}_{L(R)}^{\mu}$ and assuming they are energy independent for simplicity, we obtain for a single electron stationary current ($t \to \infty$) as follows [22]:

$$I_{k'} = 4\Gamma_{R} |t_{L,R}|^2 \left[ |f_1|^2 + |f_2|^2 + 2\text{Re}(e^{-i\phi} f_1 f_2) \right] / D^2,$$

(27)

where $\Gamma_{L(R)} = 2\pi g_{L(R)} |\tilde{t}_{L(R)}|^2$ and

$$f_{1,2} = \left( \epsilon_{L,R} - E_{2,1} \right) + \frac{i}{2} \left( \Gamma - \Gamma_{\phi} \right),$$

$$D = \left( 2\epsilon_{L,R} - E_1 - E_2 + i\Gamma \right)^2 - \varepsilon^2 - |\Gamma_{\phi}|^2.$$ 

Here $\varepsilon = E_1 - E_2$, $\Gamma = \Gamma_{L} + \Gamma_{R}$ and $\Gamma_{\phi} = \Gamma_{L} + \Gamma_{R} e^{i\phi}$.

In large-bias limit, $|\mu_{L,R} - E_{1,2}| \gg \Gamma$, the bounds of the integral $\int d\epsilon_{L,k}$ can be extended to infinity, which enables to obtain an analytic result for the total current [22]

$$I(\phi) = I_0 \frac{\varepsilon^2 + \Gamma L R \sin^2 \phi}{\varepsilon^2 + 4\Gamma L R \sin^2 \phi},$$

(28)

where $I_0 = 2\pi \Gamma L R / \Gamma$ is the resonant current in the absence of the magnetic filed. The $\phi$-dependence in Eq. (28) is an example of the Aharonov-Bohm effect. In Fig. 4, we illustrate this Aharonov-Bohm oscillations in large-bias limit, and the results for finite bias voltages by numerically integrating Eq. (27).

Now, we consider using the SCBA-ME approach to solve this same problem. To demonstrate the general algorithm, which obviously has an implication of applying to other systems, we would like to perform a direct numerical calculation, rather than making efforts to extract out any analytic results. In the noninteracting case, we apply the state basis $|0\rangle = |00\rangle$, $|1\rangle = |10\rangle$, $|2\rangle = |01\rangle$ and $|3\rangle = |11\rangle$, to convert the density operator and other operators into matrix forms. Here $|0(1)|0(1)\rangle$ corresponds to vacant (“0”) or occupied (“1”) dot state. Then, combining Eq. (13) together with its auxiliary equation (12) and Eq. (4) in the long time limit ($\dot{\rho} = 0$), we can find the steady-state solution, for both $\rho$ and $A_{\sigma \alpha \beta}$. Finally, inserting the obtained $A_{\sigma \alpha \beta}$ into Eq. (8), we are ready to calculate the steady-state current in terms of a simple trace manipulation.

Figure 4 shows a full agreement between the result from the SCBA-ME approach and the exact solution Eq. (27), for arbitrary bias voltages. This achievement desirably goes beyond the 2nd-order master equation approach, as employed in Ref. [22], where the result becomes consistent with Eq. (27) only in large bias limit. Concerning the interference pattern in Fig. 4, we would like to remark that in general the Onsager relation would lock the current (or conductance) peaks at $\phi = 2\pi n$ for any two-terminal linear response transport [24], either noninteracting or interacting. Yet, the Onsager relation cannot apply to finite voltage transport. While here in
Fig. 4 the current peaks are still at $\phi = 2\pi n$ for finite voltages, we found that this is not the case for interacting double dots. There, the phase-locking behavior as preserved in Fig. 4 breaks down.

For finite small voltages, unfortunately, either the 2nd-order master equation or the SEWF approach is no longer applicable. Now, beyond Ref. [22], we apply the SCBA-ME approach to this unexploited regime. The calculation scheme is straightforward, simply along the line of solving the noninteracting DD, as described in the previous subsection. However, there is an important issue needing care in this context. First, notice that our SCBA-ME is constructed in terms of a time-nonlocal version, see Eqs. (1) and (4), where $\rho(\tau)$ enters the integrand of $\int_0^1 d\tau \cdots$. For this time-nonlocal version of master equation, we know that even at the usual 2nd-order level, the lower bound of the integral $\int_0^\infty d\tau \cdots$ would result in a “principle-value” contribution, or, renormalization effect. For our interacting DD interferometer, as analyzed in detail in Ref. [22], the current switching behavior is caused by the occupation of a transformed dot state. On the other hand, however, we can verify that the “principle-value” integral will induce an effective coupling between the transformed dot states, $\Omega_{12} = \int_0^\infty d\epsilon_1 \epsilon_2 e^{-i(\epsilon_{1Lk} - \Delta)\tau} + \int_{-\infty}^0 d\epsilon_1 \epsilon_2 e^{i(\epsilon_{1Lk} - \Delta)\tau}$. Then, if we adopt the usual wide-band-limit model for the leads (i.e., constant density-of-states and energy-independent $t_{Lk}$), we will find that the integral can become even divergent!

This difficulty can be eliminated by applying the more realistic tunneling Hamiltonian with energy-dependent amplitudes ($t_{Lk}$). Unfortunately, to our knowledge, so far the specific behavior of $t_{Lk}$ with the energy is not well clarified. Therefore, based on certain intuition, in this work we adopt an ansatz, $|t_{Lk}|^2 = |t_0|^2 e^{-(\epsilon_{Lk} - \Delta E)^2/\sigma^2}$, where $\Delta E$ is the DD energy change associated with an electron jump, into the dots or out to the leads. Incorporating this ansatz into Eq. (1), we attach to $\Gamma^{(\pm)}(\omega)$, with a decay factor, $e^{-(\omega - E_m)^2/\sigma^2}$, where the energy difference $E_{mn} = E_m - E_n$ corresponds to the DD states $|m\rangle$ and $|n\rangle$, in association with the matrix element $\langle U(\pm \omega)|a_\mu^\dagger \rho|mn\rangle$.}

**B. Interacting Case**

When the double dots are put closer, strong Coulomb repulsion among the inter dots may lead to a reduced Hilbert space expanded only by $\{00\}$, $\{01\}$, $\{10\}$). In this large $U$ limit, as well as under large bias voltage ($U \gg |\mu_{L,R} - E_{1,2}|$), an analytic result for the steady-state current was carried out in Ref. [22]:

$$I(\phi) = I_C \frac{\varepsilon^2}{\varepsilon^2 + I_C \left(2\Gamma_R \sin^2 \frac{\phi}{2} - \varepsilon \sin \phi \right)},$$

(29)

where $I_C = 2\Gamma_L \Gamma_R / (2\Gamma_L + \Gamma_R)$ is the current in the absence of the magnetic flux. Comparing Eq. (29) with Eq. (28) for $\varepsilon \neq 0$ we find that both currents display the Aharonov–Bohm oscillations. Nevertheless, Eq. (29) shows an asymmetric behavior with respect to the magnetic flux ($\phi$). This can be verified by finding, from Eq. (29), the current peaks not at $\phi = 2\pi n$. This phase-blocking violation is an interesting consequence of Coulomb correlation in the double-path interferometer, differing from the noninteracting double dots as discussed above. More strikingly, when $\varepsilon \to 0$, Eq. (29) predicts a non-analytic current switching behavior. That is, one finds that $I = I_C$ for $\phi = 2\pi n$, but $I = 0$ for any other value of $\phi$. However, as to be shown in the following, such an unexpected switching behavior holds only for large bias voltage.

Figure 5 displays the current of the interacting DD interferometer with the magnetic flux, under finite bias voltages. We would like to mention that, for arbitrary voltages, the current pattern for $\varepsilon = 0$, aligned DD levels, will keep the current peaks at $\phi = 2\pi n$. For $\varepsilon \neq 0$, however, the current peaks will have an $\varepsilon$-dependent shift, similar as in the large bias limit. We see that, in Fig. 5, the current under finite voltage has a relatively sharp change in the vicinity of $\phi = 2\pi n$, which is an omen for the current switching in large bias limit. With the increase of the bias voltage, the switching behavior is to be more pronounced.
V. SUMMARY

We have proposed an improved master equation approach to quantum transport, by implementing a generalization from the Born to self-consistent Born approximation. Being in reasonable agreement with other scenarios of the self-consistent Born approximation, we found that the proposed scheme can give satisfactory results. For instance, it can recover not only the exact result of noninteracting transport under arbitrary voltages, but also the cotunneling as well as the challenging nonequilibrium Kondo effect in Coulomb interacting systems. Very importantly, this achievement is completely beyond the scope of the usual 2nd-order master equation under Born approximation.

As a final remark, we notice that, while the correction from the Born to self-consistent Born approximation is an efficient and well-known technique in Green’s function theory, similar correction in master equation is lacking so far among the various efforts. Since it is impossible to construct an exact and closed form of master equations in general, the scheme proposed in this work, containing the dominant contribution, is extremely desirable.

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Appendix A: Derivation of Eq. (11)

In this appendix, we present the derivation of Eq. (11), i.e., the equation-of-motion of $\hat{\rho}_j(t) \equiv U(t,\tau)[a_\mu^\dagger(t,\tau)\hat{\rho}(\tau)]$. Similar to deriving the usual 2nd-order master equation, we start with a 2nd-order expansion for the tunneling Hamiltonian $H'$, and formally obtain the same equation as Eq. (3):

$$\dot{\hat{\rho}}_j(t) = -i\mathcal{L}\hat{\rho}_j(t) - \int_0^t d\tau\text{Tr}_B[\mathcal{L}'(t)\mathcal{G}(t,\tau)\mathcal{L}'(\tau)\hat{\rho}_T(\tau)].$$

(A1)

The various superoperators in this equation have the same meaning as in Eq. (3). Explicitly, the Liouvillian self-energy superoperator can be reexpressed in Hilbert space as

$$\mathcal{L}'(t)\mathcal{G}(t,\tau)\mathcal{L}'(\tau)\hat{\rho}_T(\tau) = H'(t)G(t,\tau)H'(\tau)\hat{\rho}_T(\tau)G^\dagger(t,\tau) - G(t,\tau)H'(\tau)\rho_T(\tau)G^\dagger(t,\tau)H'(t) - H'(t)G(t,\tau)\hat{\rho}_T(\tau)H'(\tau)G^\dagger(t,\tau) - G(t,\tau)\rho_T(\tau)H'(\tau)G^\dagger(t,\tau)H'(t) \equiv I - II - III + IV.$$  

(A2)

Applying the Born approximation, $\hat{\rho}_T(\tau) \approx \rho_B\hat{\rho}_j(\tau)$, we further obtain

$$\text{Tr}_B[I] = \text{Tr}_B[H'(t)G(t,\tau)H'(\tau)\rho_B\hat{\rho}_j(\tau)G^\dagger(t,\tau)],$$

$$= \sum_{\mu\nu} \left\{ C_{\mu\nu}^{(+)}(t-\tau)a_\mu^\dagger G(t,\tau)\{a_\nu^\dagger \hat{\rho}_j(\tau)\} + C_{\mu\nu}^{(-)}(t-\tau)a_\mu^\dagger G(t,\tau)\{a_\nu \hat{\rho}_j(\tau)\} \right\},$$

(A3a)

$$\text{Tr}_B[II] = \text{Tr}_B[G(t,\tau)H'(t)\rho_B\hat{\rho}_j(\tau)G^\dagger(t,\tau)H'(t)],$$

$$= -\sum_{\mu\nu} \left\{ C_{\mu\nu}^{(-)}(t-\tau)G(t,\tau)\{a_\nu \hat{\rho}_j(\tau)\}a_\mu^\dagger + C_{\mu\nu}^{(+)}(t-\tau)G(t,\tau)\{a_\nu^\dagger \hat{\rho}_j(\tau)\} \right\},$$

(A3b)

$$\text{Tr}_B[III] = \text{Tr}_B[H'(t)G(t,\tau)\rho_B\hat{\rho}_j(\tau)H'(\tau)G^\dagger(t,\tau)],$$

$$= -\sum_{\mu\nu} \left\{ C_{\mu\nu}^{(-)}(t-\tau)a_\nu G(t,\tau)\{\hat{\rho}_j(\tau)a_\mu^\dagger\} + C_{\mu\nu}^{(+)}(t-\tau)a_\nu^\dagger G(t,\tau)\{\hat{\rho}_j(\tau)a_\mu\} \right\},$$

(A3c)

$$\text{Tr}_B[IV] = \text{Tr}_B[G(t,\tau)\rho_B\hat{\rho}_j(\tau)H'(\tau)G^\dagger(t,\tau)H'(t)],$$

$$= \sum_{\mu\nu} \left\{ C_{\mu\nu}^{(-)}(t-\tau)G(t,\tau)\{\hat{\rho}_j(\tau)a_\mu^\dagger\}a_\mu + C_{\mu\nu}^{(+)}(t-\tau)G(t,\tau)\{\hat{\rho}_j(\tau)a_\mu\}a_\mu^\dagger \right\}.$$  

(A3d)
Substituting these results into Eq. (11), a more compact notation leads to Eq. (11).

It is of crucial importance to note that, in the above $\text{Tr}_B[II]$ and $\text{Tr}_B[III]$, an extra sign change occurs when we arrange the bath operators to the two sides of $\rho_B$. We explain this more detail in the following. Consider, for instance, $\text{Tr}_B[\{a_\mu^\dagger f_\mu(\tau)\mid \rho_B f_\mu(\tau)\}]_a$. In order to utilize the cyclic property under $\text{Tr}_B[\cdots]$, i.e., $\text{Tr}_B[f_\mu(\tau)\rho_B f_\mu(\tau)] = C_{\mu\nu}^{\alpha\beta}(t - \tau)$, we have to move $f_\mu(\tau)$, crossing $\tilde{\rho}_j(\tau)$, to the right side of $\rho_B$. Since $\tilde{\rho}_j(\tau)$ contains a Fermi operator, $\alpha_j \equiv a_{\alpha_j}^\dagger$, the above movement of $f_\mu(\tau)$ will cause an additional minus sign, according to the Fermi-Dirac anticommutative relation between $a_j$ and $f_j^\dagger$. Similar consideration and result apply also to all the other terms of $\text{Tr}_B[II]$ and $\text{Tr}_B[III]$. As a consequence, the resultant extra sign of minus, very importantly, alters the commutators in the usual master equation, e.g., Eq. (3), to the

$$\Delta(\tau,\tau)[a_{\alpha_j}^\dagger \rho(\tau)].$$

Formally, from Eq. (11), we have

$$\partial_t U(t, \tau) = -i \mathcal{L} U(t, \tau) - i \sum_{\alpha, \mu, \sigma} \int \frac{d\omega}{2\pi} \{ a_{\mu}^\dagger U(t, \tau) \},$$

$$\partial_t U_{\alpha\mu}^{(\sigma)}(\omega, \tau) = -(\mathcal{L} - \sigma \omega) U_{\alpha\mu}^{(\sigma)}(\omega, \tau) - i C_{\alpha\mu}^{(\sigma)}(\omega) U(t, \tau).$$

(B3)

Here, we have further introduced

$$C_{\alpha\mu \pm}^{(\sigma)}(\omega) \hat{O} = \sum_{\nu} \{ \Gamma_{\alpha\nu}^{(\sigma)}(\omega) a_{\nu}^\dagger \hat{O} \pm \Gamma_{\alpha\nu}^{(\sigma)}(\omega) \hat{O} a_{\nu}^\dagger \},$$

(B4)

and

$$U_{\alpha\mu}^{(\sigma)}(\omega, \tau) = -i \int_{t_0}^\tau d\tau' e^{-i(\mathcal{L} - \sigma \omega)(\tau - \tau')} C_{\alpha\mu}^{(\sigma)}(\omega) U(\tau, \tau_0).$$

(B5)

With these identifications, Eq. (6) can be recast to the following set of coupled equations:

$$\dot{\rho}(t) = -i \mathcal{L} \rho(t) - i \sum_{\alpha, \mu, \sigma} \int \frac{d\omega}{2\pi} \{ a_{\mu}^\dagger \phi_{\alpha\mu}^{(\sigma)}(\omega, t) \},$$

(B6a)

$$\dot{\phi}_{\alpha\mu}^{(\sigma)}(\omega, t) = -i (\mathcal{L} - \sigma \omega) \phi_{\alpha\mu}^{(\sigma)}(\omega, t) - i C_{\alpha\mu}^{(\sigma)}(\omega) \rho(t)$$

$$- i \sum_{\alpha', \mu', \sigma'} \int d\omega' \{ a_{\mu'}^\dagger \phi_{\alpha'\mu'}^{(\sigma)}(\omega', t) \},$$

(B6b)

$$\dot{\phi}_{\alpha'\mu'}^{(\sigma)}(\omega, \omega', t) = -i (\mathcal{L} - \sigma \omega - \sigma' \omega') \phi_{\alpha'\mu'}^{(\sigma)}(\omega, \omega', t)$$

$$- i C_{\alpha'\mu'}^{(\sigma)}(\omega') \phi_{\alpha'\mu'}^{(\sigma)}(\omega, t).$$

(B6c)

Eqs. (B4), favorably, recover the HMEs truncated until the second tier ($n_{\text{tier}} = 2$) [13], up to an unimportant difference in Eq. (B6a). That is, the HMEs with a cutoff at $n_{\text{tier}} = 2$ has one more term, $C_{\alpha\mu}^{(\sigma)}(\omega) \phi_{\alpha\mu}^{(\sigma)}(\omega', t)$. We have numerically examined, however, that this term has negligible effect.

In good agreement with other SCBA scenarios in broad physical contexts, we find that the SCBA-ME can give satisfactory results, not only for noninteracting transports, but also for highly correlated processes. For instance, it can predict the nonequilibrium Kondo effect [5], while this challenging effect is completely beyond the scope of the second-order master equation, as well as some improved approach [10]. We have also checked that the proposed SCBA-ME contains all the cotunneling processes in Coulomb blockade systems [10]. On the other hand, in applying the HMEs, it was found that the dominant contributions are usually obtained by a cutoff at $n_{\text{tier}} = 2$, while the pure numerical convergence may be achieved by including more tiers of the HMEs, at a price of much more expensive computations. The above facts, combined together, then suggest that the SCBA-ME is a reliable quantum transport approach. Therefore, based on the SCBA-ME, the above identification of Eqs. (B4) actually helps us extract out the dominant tiers. That is, the first two tiers of the HMEs would dominate the numerical contribution, as well as the underlying physics.
Appendix C: Lorentzian Spectrum

In most cases for a system coupled to a continuum, a Lorentzian type of spectral density function is more reasonable than a constant. In quantum transport, for coupling with the leads, we therefore assume

\[ \Gamma_{\alpha \mu \nu}(\omega) = \frac{\Gamma_{\alpha \mu \nu} W_\alpha^2}{(\omega - \mu_\alpha)^2 + W_\alpha^2} \]  

(C1)

In certain sense, this form corresponds to a half-occupied conduction band for each lead, which fixes the center of the Lorentzian function at the Fermi level (chemical potential), \( \mu_\alpha \), of the lead. In Eq. (C1), \( W_\alpha \) characterizes the bandwidth of the \( \alpha \)-th lead. Quite naturally, the constant coupling rate can be recovered from Eq. (C1) by assuming \( W_\alpha \rightarrow \infty \), yielding \( \Gamma_{\alpha \mu \nu}(\omega) = \Gamma_{\alpha \mu \nu} \).

Corresponding to the Lorentzian spectral density Eq. (C1), the correlation function of Eq. (14) can be expressed as

\[ C_{\alpha \mu \nu}(\omega) = \frac{1}{2} \left[ \Gamma_{\alpha \mu \nu}(\mp \omega) + i \Lambda_{\alpha \mu \nu}(\mp \omega) \right]. \]  

(C2)

The second quantity is related to the first one through the well-known dispersive relation as

\[ \Lambda_{\alpha \mu \nu}(\omega) = \mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \frac{1}{\omega - \omega'} \Gamma_{\alpha \mu \nu}(\omega') \]

\[ = \frac{\Gamma_{\alpha \mu \nu}}{\pi} \left\{ \text{Re} \left[ \Psi \left( \frac{1}{2} + i \frac{\omega_\alpha}{\pi} \right) \right] + \omega_\alpha \right\} \]  

(C3)

where \( \mathcal{P} \) denotes the principle value of the integral, and \( \Psi(x) \) is the digamma function.