Number-resolved master equation approach to quantum transport under the self-consistent Born approximation

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We construct a particle-number(n)-resolved master equation (ME) approach under the self-consistent Born approximation (SCBA) for quantum transport through mesoscopic systems. The formulation is essentially non-Markovian and incorporates the interlay of the multi-tunneling processes and many-body correlations. The proposed n-SCBA-ME goes completely beyond the scope of the Born-Markov master equation, being applicable to transport under small bias voltage, in non-Markovian regime and with strong Coulomb correlations. For steady state, it can recover not only the exact result of noninteracting transport under arbitrary voltages, but also the challenging nonequilibrium Kondo effect. Moreover, the n-SCBA-ME approach is efficient for the study of shot noise. We demonstrate the application by a couple of representative examples, including particularly the nonequilibrium Kondo system.

Master equation, quantum transport, shot noise spectrum

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I. INTRODUCTION

In addition to the Landauer-Büttiker scattering theory and the non-equilibrium Green’s function method [1, 2], as an alternative choice, the rate or master equation approach is very convenient for transport through nanostuctures with a few discrete states [3–12]. Moreover, the number(n)-resolved version of the transport master equation [6, 7, 11–13], has been demonstrated as an efficient scheme for studies of shot noise and counting statistics in mesoscopic transports, including also the large-derivation analysis [14].

However, the perturbative master equation is usually up to the 2nd-order expansion of the tunneling Hamiltonian, which makes it applicable only in the limit of large bias voltage. This 2nd-order master equation (2nd-BA) does not account for the level’s broadening effect. Moreover, if applying to Coulomb interacting system, it cannot describe cotunneling process and the nonequilibrium Kondo effect. Therefore, higher-order expansions of the tunneling Hamiltonian are necessary sometimes, as the efforts made in literature [8–10, 15, 16].

In this work, by an insight from the Green’s function theory, we generalize the master equation approach from the usual 2nd-order Born approximation (BA) to self-consistent Born approximation (SCBA). We will demonstrate that the effect of this improvement is remarkable: it can recover not only the exact result of noninteracting transport under arbitrary voltages, but also the nonequilibrium Kondo effect in Coulomb interacting system. In particular, the particle-number(n)-resolved version of the SCBA-ME (n-SCBA-ME) provides an efficient scheme for studying the shot noise and counting statistics, as to be illustrated by a couple of application examples in this work.

The paper is organized as follows. In Sec. II we outline the main formulation of the SCBA-ME, where the steady state current and an illustrative example will be presented. In Sec. III, we continue the formal construction of the n-SCBA-ME and provide the calculation scheme of noise spectrum, while leaving the specific examples in Sec. IV with in particular the shot noise of the nonequilibrium Kondo system. Finally, we summarize the work in Sec. V.

II. FORMULATION OF THE SCBA-ME

In general we describe a transport setup by $H = H_S(a^\dagger_\mu, a_\mu) + H_{res} + H'$. Here $H_S$ is the Hamiltonian of the central system embedded between two leads, with $a^\dagger_\mu$ ($a_\mu$) the creation (annihilation) operator of the state $|\mu\rangle$. The other two Hamiltonians, $H_{res}$ and $H'$, describe the leads and their tunnel coupling to the central system. They are modeled by, respectively, $H_{res} = \sum_{\alpha=L,R} \sum_k \epsilon_{\alpha k} b^\dagger_{\alpha k} b_{\alpha k}$ and $H' = \sum_{\alpha=L,R} \sum_{\mu k} (t_{\alpha\mu} a^\dagger_{\mu k} b_{\alpha k} + \text{H.c.})$ with $b^\dagger_{\alpha k}$ ($b_{\alpha k}$) the creation (annihilation) operator of electron in state $|k\rangle$ of the left ($L$) and right ($R$) leads.
A. ME under Born Approximation

In a compact form, the master equation under the Born approximation can be expressed as [12]

$$\dot{\rho}(t) = -i\mathcal{L}\rho(t) - \sum_{\mu\sigma} \left\{ [\sigma^\dagger_{\mu}, A^{\sigma}_{\mu\nu}(t)] + \text{H.c.} \right\}. \quad (1)$$

In this work we use a reduced system of units by setting \( \hbar = k_B = e = 1 \) for the Planck constant, the Boltzmann constant and the electron charge. In Eq. (1) we also define: \( \sigma = + \) and \( - \), \( \bar{\sigma} = -\sigma \), \( a^+_\mu = a^\dagger_\mu \), and \( a^-\mu = a_\mu \).

The superoperators read \( \mathcal{L}\rho = [H^\dagger, \rho] \), and \( A^{\sigma}_{\mu\nu}(t) = \sum_{\alpha=L,R} A^{\sigma}_{\alpha\mu\nu}(t) \) while \( A^{(+)}_{\alpha\mu\nu}(t) = \sum_{\nu} \int_t^\infty d\tau C^{(+)}_{\alpha\mu\nu}(t - \tau) \{ G(t, \tau) [a^+_\alpha, \rho(\tau)] \} \). \( G(t, \tau) \) is the free propagator, determined by the system Hamiltonian as \( G(t, \tau) = e^{-i\mathcal{L}(t-\tau)} \).

For the convenience of later use, we present a specific characterization for \( C^{(+)}_{\alpha\mu\nu}(t - \tau) \), the correlation function of the reservoir electrons (in local equilibrium):

$$C^{(+)}_{\alpha\mu\nu}(t - \tau) = \langle f^{(+)}_{\alpha\mu}(t)f^{(+)}_{\alpha\nu}(\tau) \rangle_B. \quad (2)$$

Here, \( f^{(+)}_{\alpha\mu}(t) = f_{\alpha\mu}^+(t) \) and \( f^{(-)}_{\alpha\mu}(t) = f_{\alpha\mu}(t) \), via rewriting the tunneling Hamiltonian as \( H^\dagger = \sum_{\alpha=L,R} \sum_\mu \sum_\sigma (a^\dagger_{\alpha\mu} f_{\alpha\mu} + \text{H.c.}) \) by introducing \( f_{\alpha\mu} = \sum_k t_{\alpha\mu k} a_{\mu k} \). The time dependence of the operators in \( C^{(+)}_{\alpha\mu\nu}(t - \tau) \) originates from the interaction picture with respect to the reservoir Hamiltonian, and the average \langle \cdot \cdot \cdot \rangle_B is over the reservoir states. Moreover, we introduce the Fourier transform of \( C^{(+)}_{\alpha\mu\nu}(t - \tau) \):

$$C^{(+)}_{\alpha\mu\nu}(t - \tau) = \int_{-\infty}^\infty \frac{d\omega}{2\pi} e^{\pm i\omega(t-\tau)} \Gamma^{(+)}_{\alpha\mu\nu}(\omega). \quad (3)$$

Accordingly, we have \( \Gamma^{(+)}_{\alpha\mu\nu}(\omega) = \Gamma_{\alpha\mu\nu}(\omega) n_{\alpha}^{(+)}(\omega) \) and \( \Gamma^{(-)}_{\alpha\mu\nu}(\omega) = \Gamma_{\alpha\mu\nu}(\omega) n_{\alpha}^{(-)}(\omega) \), where \( \Gamma_{\alpha\mu\nu}(\omega) = 2\pi \sum_k t_{\alpha\mu k}^2 \pi(\omega - \epsilon_k) \delta(\omega - \epsilon_k) \) is the spectral density function of the reservoir \( \alpha \), \( n_{\alpha}^{(+)}(\omega) \) denotes the Fermi function \( n_{\alpha}(\omega) \), and \( n_{\alpha}^{(-)}(\omega) = 1 - n_{\alpha}(\omega) \) is introduced for brevity. Alternatively, we may introduce as well the Laplace transform of \( C^{(+)}_{\alpha\mu\nu}(t - \tau) \), denoting by \( C^{(+)}_{\alpha\mu\nu}(\omega) \), which is related with \( \Gamma^{(+)}_{\alpha\mu\nu}(\omega) \) through the well known dispersive relation:

$$C^{(+)}_{\alpha\mu\nu}(\omega) = \frac{\Gamma_{\alpha\mu\nu}\pi}{2\omega} \left[ i \omega + i\omega' + i\omega'' + \bar{\sigma} \right] \Gamma^{(+)}_{\alpha\mu\nu}(\omega'). \quad (4)$$

In this work, for the reservoir spectral density function, we assume a Lorentzian form as

$$\Gamma_{\alpha\mu\nu}(\omega) = \frac{\Gamma_{\alpha\mu\nu} W^2_{\alpha}}{(\omega - \mu_{\alpha})^2 + W^2_{\alpha}}, \quad (5)$$

In some sense, this assumption corresponds to a half-occupied band for each lead, which peaks the Lorentzian center at the chemical potential \( \mu_{\alpha} \). \( W_{\alpha} \) characterizes the bandwidth of the \( \alpha \)th lead. Obviously, the usual constant spectral density function is recovered from Eq. (5) in the limit \( W_{\alpha} \rightarrow \infty \), yielding \( \Gamma_{\alpha\mu\nu}(\omega) = \Gamma_{\alpha\mu\nu} \). Corresponding to the above Lorentzian spectral density function, straightforwardly, we obtain

$$C^{(+)}_{\alpha\mu\nu}(\omega) = \frac{1}{2} \left[ \Gamma^{(\pm)}_{\alpha\mu\nu}(\mp \omega) + i\Lambda^{(+)}_{\alpha\mu\nu}(\mp \omega) \right]. \quad (6)$$

The imaginary part, through the dispersive relation, is associated with the real one as

$$\Lambda^{(+)}_{\alpha\mu\nu}(\omega) = \frac{P}{\pi} \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \frac{1}{\omega' - \omega} \Gamma^{(+)}_{\alpha\mu\nu}(\omega),$$

where \( P \) stands for the principle value and \( \Psi(x) \) is the digamma function.

We remark that the 2nd-order master equation can apply only to transport under large bias voltage. That is, the Fermi levels of the leads should be considerably away from the system levels, by at least several times of the level’s broadening.

B. ME under Self-Consistent Born Approximation

The basic idea to improve the 2nd-ME can follow what is typically done in the Green’s function theory, i.e., correcting the self-energy diagram from the Born to a self-consistent Born approximation. In our case, the SCBA scheme can be implemented by replacing the free propagator in the 2nd-order master equation, \( G(t, \tau) = e^{-i\mathcal{L}(t-\tau)} \), by an effective one, \( \mathcal{U}(t, \tau) \), which propagates a state with the precision of the 2nd-order Born approximation. From this type of consideration, the generalized SCBA-ME follows Eq. (1) directly as [17]:

$$\dot{\rho}(t) = -i\mathcal{L}\rho(t) - \sum_{\mu\sigma} \left\{ [\sigma^\dagger_{\mu}, A^{\sigma}_{\mu\nu}(t)] + \text{H.c.} \right\}. \quad (8)$$

Here \( A^{\sigma}_{\mu\nu}(t) = \sum_{\alpha=L,R} A^{\sigma}_{\alpha\mu\nu}(t) \), and \( A^{\sigma}_{\alpha\mu\nu}(t) = \sum_{\nu} \int_t^\infty d\tau C^{(A)}_{\alpha\mu\nu}(t - \tau) \{ \mathcal{U}(t, \tau) [a^\dagger_{\alpha\nu}, \rho(\tau)] \} \). To close this master equation, let us define \( \tilde{\rho}_j(t) \equiv \mathcal{U}(t, \tau) [a^\dagger_{j\alpha}, \rho(\tau)] \) (here and in the following we use \( \tilde{\cdot} \) to denote the double indices \((\nu, \sigma)\) for the sake of brevity). Then, the equation-of-motion (EOM) of this auxiliary object reads

$$\dot{\tilde{\rho}}_j(t) = -i\mathcal{L}\tilde{\rho}_j(t) - \int_t^\infty dt' \Sigma^{(A)}_{\alpha\nu}(t - t') \tilde{\rho}_j(t'). \quad (9)$$

In this equation the 2nd-order self-energy superoperator, \( \Sigma^{(A)}_{\alpha\nu}(t - t') \), differs from the usual one because it involves anticommutators, rather than the commutators.
in the 2nd-order master equation. More explicitly, we have
\[
\int_\tau^t dt' \Sigma_2^{(A)}(t - t') \rho_j(t') = \sum_{\mu} \left[ \{ \rho_{\mu}, A_{\mu}^{(t)} \} + \{ \rho_{\mu}, A_{\mu}^{(t)} \} + \{ a_{\mu}, A_{\mu}^{(t)} \} \right] + \{ a_{\mu}, A_{\mu}^{(t)} \}, \quad (10)
\]
where \( A_{\mu}^{(t)} \) is defined as \( A_{\mu}^{(t)} = \sum_{\alpha=L,R} \int_\tau^t dt' C^{(\alpha)}(t - t') \left\{ e^{-i \epsilon_{\alpha}(t - t')} [a_{\alpha}^\dagger \rho_j(t')] \right\} \).

Because of the anticommutative brackets here, we stress that the propagation of \( \rho_j(t) \) does not satisfy the usual 2nd-order master equation. This, in certain sense, violates the so-called quantum regression theorem.

**C. Steady State Current**

Within the framework of SCBA-ME, similar to its 2nd-order counterpart, current through the \( o \)th lead reads
\[
I_{o}(t) = 2 \sum_{\mu} \text{Re} \left\{ \text{Tr} \left[ A_{\alpha \mu}^{(t)}(t) a_{\mu} - A_{\alpha \mu}^{(t)}(t) a_{\mu}^\dagger \right] \right\}. \quad (11)
\]
For steady state, consider the integral \( \int_0^\infty d\tau [\cdots] \rho(\tau) \) in \( A_{\alpha \mu}^{(t)}(t) \). Since physically, the correlation function \( C^{(\alpha)}_{\mu \nu}(t - \tau) \) in the integrand is nonzero only on finite timescale, we can replace \( \rho(\tau) \) in the integrand by the steady state \( \tilde{\rho} \), in the long time limit \( (t \rightarrow \infty) \). After this replacement, we obtain
\[
A_{\alpha \mu}^{(t)} = \sum_{\nu} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Gamma_{\alpha \mu}(\omega) \left\{ \{ \pm \omega \} [a_{\alpha}^\dagger \tilde{\rho}] \right\}. \quad (12)
\]
Then, substituting this result into Eq. (8), we can straightforwardly solve for \( \tilde{\rho} \) and calculate the steady state current.

Based on \( \tilde{\rho} \), to obtain further the current, we first introduce \( \Phi_{1,2}(\omega) = \text{Tr} \left[ a_{\mu} \rho_{1,2}(\omega) \right] \) and \( \Phi_{1,2}(\omega) = \text{Tr} \left[ a_{\mu} \rho_{1,2}(\omega) \right] \) using Eq. (9), with an initial condition of \( \rho_{1,2}(0) = \tilde{\rho}_{2,0} \) and \( \tilde{\rho}_{2,0}(0) = a_{\mu}^\dagger \tilde{\rho} \). To simplify notations, we denote the various matrices in boldface form: \( \Phi_{1}(\omega), \Phi_{2}(\omega) \) and \( \Gamma_{L,R} \). Now, if \( \Gamma_{L} \) is proportional to \( \Gamma_{R} \) by a constant, the steady state current can be recast to the Landauer-Büttiker type:
\[
I = 2 \text{Re} \left\{ \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} [n_L(\omega) - n_R(\omega)] T(\omega) \right\}, \quad (13)
\]
where tunneling coefficient, very compactly, is given by
\[
T(\omega) = \text{Tr} \left[ \Gamma_{L} \Gamma_{R} (\Gamma_{L}^\dagger + \Gamma_{R})^{-1} \{ \Phi(\omega) \} \right]. \quad (14)
\]
Here \( \Phi(\omega) = \Phi_{1}(\omega) + \Phi_{2}(\omega) \).

Now we demonstrate that, for a noninteracting system, the above stationary current coincides precisely with the nonequilibrium Green’s function approach, both giving the exact result under arbitrary bias voltage. In general, a noninteracting system can be described by \( \hat{H}_S = \sum_{\mu} h_{\mu} a_{\mu}^\dagger a_{\mu} \). Straightforwardly, we obtain the EOM for \( \varphi \) as follows:
\[
-i\omega \varphi_i(\omega) - \varphi_i(0) = -i \hat{\mathbf{H}} \Phi_i(\omega) - i \Sigma(\omega) \varphi_i(\omega). \quad (15)
\]
where \( \varphi_i(0) \) stand for the initial conditions, \( \varphi_{1,1}(0) = \text{Tr} \left[ a_{\mu} a_{\mu}^\dagger \right] \) and \( \varphi_{2,2}(0) = \text{Tr} \left[ a_{\mu} a_{\mu}^\dagger \right] \). The tunnel-coupling self-energy \( \Sigma(\omega) \) reads \( \Sigma(\omega) = -i \sum_{\alpha} \left[ C_{\alpha \mu}(\omega) + C_{\alpha \mu}^\dagger(-\omega) \right] \), or
\[
\Sigma(\omega) = \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \Gamma_{\alpha \mu}(\omega) \quad (16)
\]
Then, based on Eq. (15), summing up \( \varphi_{1}(\omega) \) and \( \varphi_{2}(\omega) \) yields
\[
\Phi(\omega) = i \left[ \omega - \mathbf{H} - \Sigma(\omega) \right]^{-1} \quad (17)
\]
In deriving this result, the cyclic property under trace and the anti-commutator, \( \{ a_{\mu}, a_{\mu}^\dagger \} = \delta_{\mu \nu}, \) have been used. Eq. (17) is nothing but the exact Green’s function for transport through a noninteracting system, giving thus the exact stationary current after inserting it into the above current formula.

**D. Interacting Case**

To show the application of the proposed SCBA-ME to interacting system, as an illustrative example, we consider the transport through an interacting quantum dot described as
\[
\hat{H}_S = \sum_{\mu} \left( \epsilon_{\mu} a_{\mu}^\dagger a_{\mu} + \frac{U}{2} n_{\mu} \right). \quad (18)
\]
Here 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 \( U n_{\uparrow} n_{\downarrow} \) is the number operator and \( U \) represents the interacting strength.

First, we note that \( C_{\alpha \mu}^{(\pm)} \) is diagonal with respect to the spin states, i.e., \( C_{\alpha \mu}^{(\pm)}(t) = \delta_{\mu \nu} C_{\alpha \nu}^{(\pm)}(t) \) and \( \gamma_{\alpha \mu}^{(\pm)} = \Gamma_{\alpha \mu}^{(\pm)} \). Then, we specify the states involved in the transport as \( |0\rangle, |\uparrow\rangle, |\downarrow\rangle \) and \( |\bar{\mu}\rangle \), corresponding to the empty, spin-up, spin-down and double occupancy states, respectively. Using this basis, we reexpress the electron operator in terms of projection operator, \( a_{\mu}^\dagger = |\mu\rangle \langle 0| + (-1)^{\mu} |\bar{\mu}\rangle \langle \bar{\mu}|, \) where the convention \((-1)^{\uparrow} = 1 \) and


\[ (-1)^k = -1 \text{ is implied. For a solution of the steady state, we have} \]

\[ A^{(\pm)}_{\alpha \mu \rho} = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Gamma^{(\pm)}_{\alpha \mu \rho}(\omega)U(\pm \omega)[a^\pm_{\mu \rho}] . \] \[ (19) \]

Straightforwardly, after some algebra, \( U(\pm \omega)[a^\pm_{\mu \rho}] \) can be carried out as

\[ U(\omega)[a^+_{\mu \rho}] = [\lambda^+_{\mu}(\omega)|0\rangle + \kappa^+_{\mu}(\omega)(-1)^n|d\rangle] , \]

\[ U(-\omega)[a_{\mu \rho}] = [\lambda^-_{\mu}(\omega)|0\rangle(\mu) + \kappa^-_{\mu}(\omega)(-1)^n|\bar{\mu}\rangle] , \] \[ (20) \]

where

\[ \lambda^+_{\mu}(\omega) = i \frac{\Pi^{-1}_{1\mu}(\omega)\bar{\rho}_{00} - \Sigma^-_{\mu}(\omega)\bar{\rho}_{\mu\bar{\mu}}}{\Pi^{-1}_{1\mu}(\omega)\Pi^{-1}_{1\mu}(\omega)} , \]

\[ \lambda^-_{\mu}(\omega) = i \frac{\Pi^{-1}_{1\mu}(\omega)\rho_{\mu\bar{\mu}} - \Sigma^-_{\mu}(\omega)\rho_{dd}}{\Pi^{-1}_{1\mu}(\omega)\Pi^{-1}_{1\mu}(\omega)} , \]

\[ \kappa^+_{\mu}(\omega) = i \frac{-\Sigma^+_{\bar{\mu}}(\omega)\bar{\rho}_{00} + \Pi^{-1}_{1\mu}(\omega)\bar{\rho}_{\mu\bar{\mu}}}{\Pi^{-1}_{1\mu}(\omega)\Pi^{-1}_{1\mu}(\omega)} , \]

\[ \kappa^-_{\mu}(\omega) = i \frac{-\Sigma^+_{\bar{\mu}}(\omega)\rho_{\mu\bar{\mu}} + \Pi^{-1}_{1\mu}(\omega)\rho_{dd}}{\Pi^{-1}_{1\mu}(\omega)\Pi^{-1}_{1\mu}(\omega)} . \]

Here, we introduced \( \Pi^{-1}_{1\mu}(\omega) = \omega - \epsilon_{\mu} - \Sigma_{0\mu}(\omega) - \Sigma^+_{\mu}(\omega) \), and \( \Pi^+_{1\mu}(\omega) = \omega - \epsilon_{\mu} + \Sigma_{0\mu}(\omega) - \Sigma^-_{\mu}(\omega) \). The self-energies \( \Sigma_{0\mu}(\omega) \) and \( \Sigma^\pm_{\mu}(\omega) \) are given by

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

\[ \Sigma^\pm_{\mu}(\omega) = \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \frac{\Gamma^{(\pm)}_{\mu}(\omega')}{\omega' - \omega - \epsilon_{\bar{\mu}} + \epsilon_{\mu} - \omega' + i 0^+} + \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \frac{\Gamma^{(\pm)}_{\mu}(\omega')}{\omega' - \omega - E_d + \omega' + i 0^+} . \] \[ (21) \]

Then, we find the solution of \( \varphi(\omega) \) as

\[ \varphi(\omega) = i(1 - n_{\bar{\mu}}) \frac{\omega - \epsilon_{\mu} - \Sigma_{0\mu} + U\Sigma^+_{\mu}(\omega - \epsilon_{\mu} - U - \Sigma_{0\mu} - \Sigma^+_{\mu})^{-1} + \frac{i n_{\bar{\mu}}}{\omega - \epsilon_{\mu} - U - \Sigma_{0\mu} - U\Sigma^+_{\mu}(\omega - \epsilon_{\mu} - \Sigma_{0\mu} - \Sigma^+_{\mu})^{-1}} , \] \[ (22) \]

where \( n_{\mu} = \rho_{\mu\bar{\mu}} + \rho_{\mu\mu} \), and \( 1 - n_{\mu} = \rho_{\mu\bar{\mu}} + \rho_{00} \). This result, precisely, coincides with the one from the EOM technique of the nonequilibrium Green’s function (nGF) [2]. Therefore, as discussed in detail in the book by Haug and Jauho [2], this solution contains the remarkable nonequilibrium Kondo effect.

At high temperatures, the terms \( U\Sigma^\pm_{\mu}(\cdots)^{-1} \) vanish, reducing thus Eq. (22) to

\[ \varphi_{HF}(\omega) = \frac{i(1 - n_{\bar{\mu}})}{\omega - \epsilon_{\mu} - \Sigma_{0\mu}} + \frac{i n_{\bar{\mu}}}{\omega - \epsilon_{\mu} - U - \Sigma_{0\mu}} . \] \[ (23) \]

Here we use \( \varphi_{HF} \) to imply the result at the level of a mean-field Hartree-Fock approximation. Actually, Eq. (23) can also be derived from the EOM technique of nGF at lower-order cutoff, by using a mean-field approximation [2]. The point is that, noting the broadening effect contained, even this simple result goes beyond the scope of the 2nd-order master equation. In Fig. 1 we plot the current-voltage relation based on Eq. (22) against that from Eq. (23).

![Coulomb staircase in the current-voltage curve. Inset: the corresponding differential conductance. The result based on Eq. (22) is plotted against the Hartree-Fock (HF) solution Eq. (23). Parameters: \( \Gamma_L = \Gamma_R = \Gamma/2, \epsilon_0 = 7\Gamma, U = 10\Gamma, \) and \( k_B T = 0.1\Gamma \). The bias voltage is set to \( \mu_L = -\mu_R = eV/2 \) which assumes the zero-bias Fermi level as energy reference. In this work (here and in other figures below) we use a reduced system of units by assuming \( h = e = k_B = 1 \), and setting \( \Gamma = 1 \) for an arbitrary unit of energy.]

III. FORMULATION OF THE n-SCBA-ME

Now we proceed to construct the particle number (“n”) resolved SCBA-ME, along the same line in constructing the “n”-resolved 2nd-order master equation [11, 12]. The basic idea is to split the Hilbert space of the reservoirs into a set of subspaces, each labeled by \( n \). Then, do the average (trace) over each subspace and define the corresponding reduced density matrix as \( \rho^{(n)}(t) \). To be specific, consider the \( \rho^{(n)}(t) \) conditioned on the electron number arrived to the right lead, which obeys

\[ \rho^{(n)} = -i \mathcal{L} \rho^{(n)} - \sum_{\mu} \left\{ a^\dagger_{\mu} A_{\mu\bar{\mu}}^{(n)}(t) + a_{\mu} A_{\mu\bar{\mu}}^{(n)}(t) + A_{\mu\bar{\mu}}^{(n)}(t) a^\dagger_{\mu} - A_{\mu\bar{\mu}}^{(n)}(t) a_{\mu} + A_{\mu\bar{\mu}}^{(n)}(t) a^\dagger_{\mu} + A_{\mu\bar{\mu}}^{(n)}(t) a_{\mu} \right\} . \] \[ (24) \]

Here \( A_{\mu\bar{\mu}}^{(n)}(t) = \sum_{\nu} \int_0^\infty d\tau C_{\nu\mu}(t - \tau) \rho^{(n)}_{j}(t, \tau) \), while the summation over \( \nu \) makes sense in regard to the abbreviation \( j = \{ \nu, \sigma \} \). In Eq. (24), the appearing of \( \rho^{(n\pm)}(t, \tau) \) is owing to a more tunneling event (forward/backward) involved in the process.
of the corresponding terms. In particular, $\tilde{\rho}_j^{(n)}(t, \tau)$ is the $n$-dependent version of the quantity $\tilde{\rho}_j(t, \tau) = U(t, \tau)\rho(\tau)$, satisfying an OME according to Eq. (9):

$$\tilde{\rho}_j^{(n)} = -i\mathcal{L}\tilde{\rho}_j^{(n)} - \sum_{\mu} \left\{ [a_{\mu} A^{(-)}_{\mu\rho j}^{(n)} + a_{\mu} A^{(+)\rho j}_{\mu\rho}^{(n)} + A^{(-)}_{\mu\rho j}^{(n)}, a_{\mu}^{\dagger} + A^{(-)}_{\mu\rho j}^{(n)}, a_{\mu}] + \text{H.c.} \right\}. \tag{25}$$

In this equation we introduced $A^{(\sigma)^{\prime}}_{\alpha\mu\nu}(t) = \sum_{\nu'} \int_t^{t'} dt' C^{(\sigma)^{\prime}}_{\alpha\mu\nu}(t-t') \{ e^{-i\mathcal{L}(t-t')} [a_{\nu'} \rho_j^{(n)}(t')] \}$.

The $n$-resolved master equation contains rich information. This allows a great variety of its applications, including such as a convenient calculation of shot noise spectrum and the study of full counting statistics [14]. In the remaining part of this work we focus on the issue of shot noise spectrum. The noise spectrum, $S(\omega)$, is the Fourier transform of the current correlation function $S(t) = \langle I(t)I(0) \rangle_s$, defined in the steady state. Very conveniently, within the framework of the n-ME, one can calculate $S(\omega)$ by using the MacDonald’s formula [11]: $S(\omega) = 2\omega \int_0^{\infty} dt \sin(\omega t) \frac{d}{dT} \langle n^2(t) \rangle$, where $\langle n^2(t) \rangle = \sum_n n^2 P(n, t) = Tr \sum_n n^2 \rho^{(n)}(t)$, and the $n$-counting starts with the steady state (\bar{\rho}). Based on Eq. (24), one can express $\frac{d}{dT} \langle n^2(t) \rangle$ in terms of $A^{(\sigma)^{\prime}}_{\alpha\mu\nu}(t)$ and $A^{(\sigma)^{\prime}}_{\alpha\mu\nu}(t)$. The former has been introduced in Eq. (8), needing only to replace $\rho(\tau)$ by $\bar{\rho}$. The latter reads $A^{(\sigma)^{\prime}}_{\alpha\mu\nu}(t) = \sum_{\nu'} \int_t^{t'} dt' C^{(\sigma)^{\prime}}_{\alpha\mu\nu}(t-t') [\tilde{N}_j(t, \tau)]$, where $\tilde{N}_j(t, \tau) = \sum_n n \tilde{\rho}_j^{(n)}(t, \tau)$. Then, the MacDonald’s formula becomes

$$S(\omega) = 2\omega \text{Im} \sum_{\mu} \text{Tr} \left\{ 2 \left[ A^{(-)}_{\mu\rho j}(\omega) a_{\mu} - A^{(+)\rho j}_{\mu\rho}^{(n)}(\omega) a_{\mu}^\dagger \right] + \left[ A^{(-)}_{\mu\rho j}(\omega) a_{\mu}^\dagger + A^{(+)\rho j}_{\mu\rho}^{(n)}(\omega) a_{\mu} \right] \right\}. \tag{26}$$

This result is obtained after Laplace transforming $A^{(\sigma)^{\prime}}_{\alpha\mu\nu}(t)$ and $A^{(\sigma)^{\prime}}_{\alpha\mu\nu}(t)$. More explicitly,

$$A^{(\sigma)^{\prime}}_{\alpha\mu\nu}(\omega) = \sum_{\nu'} \int_0^{\infty} \frac{d\omega'}{2\pi} \Gamma^{(\sigma)^{\prime}}_{\alpha\mu\nu}(\omega') \tilde{U}(\omega + \sigma\omega') [a_{\nu'} \bar{\rho}(\omega)],$$

where the Laplace transformation of the steady state reads $\tilde{\rho}(\omega) = i\bar{\rho}/\omega$, and the propagator $\tilde{U}$ in frequency domain is defined through Eq. (9). On the other hand, $A^{(\sigma)^{\prime}}_{\alpha\mu\nu}(\omega)$ reads

$$A^{(\sigma)^{\prime}}_{\alpha\mu\nu}(\omega) = \sum_{\nu'} \int_0^{\infty} \frac{d\omega'}{2\pi} \Gamma^{(\sigma)^{\prime}}_{\alpha\mu\nu}(\omega') \tilde{U}(\omega + \sigma\omega') [a_{\nu'} \bar{N}(\omega)].$$

In deriving this result, we introduced an additional propagator through $\tilde{N}_j(t, \tau) = \tilde{U}(t-\tau)\tilde{N}_j(\tau)$, with $\tilde{N}_j(\tau) = a_{\nu}^{\dagger} \bar{N}(\tau)$ as the initial condition which is defined by $N(\tau) = \sum_n n \rho^{(n)}(\tau)$. $\tilde{U}(\omega)$ and $N(\omega)$ can be obtained via Laplace transforming the following EOMs. (i) For $N(\omega)$, based on the n-SCBA-ME we obtain

$$\tilde{N}(t) = -i\mathcal{L}N(t) - \sum_{\mu} \left\{ [a_{\mu}, A^{(\sigma)^{\prime}}_{\mu\nu}(t)] + \text{H.c.} \right\} + \sum_{\mu} \left\{ [A^{(-)}_{\mu\rho j} a_{\mu} - A^{(+)\rho j}_{\mu\rho}^{(n)}(t)] + \text{H.c.} \right\}. \tag{27}$$

(ii) For $\tilde{U}(\omega)$, from Eq. (25) we have

$$\tilde{N}_j(t) = -i\mathcal{L}\tilde{N}_j(t) - \int_{\tau}^{t} dt' \Sigma_{\alpha\mu\nu}^{(A)}(t-t') \tilde{N}_j(t') - \sum_{\mu} \left\{ [A^{(-)}_{\mu\rho j} a_{\mu} - A^{(+)\rho j}_{\mu\rho}^{(n)}(t)] + \text{H.c.} \right\}. \tag{28}$$

The self-energy superoperator $\Sigma_{\alpha\mu\nu}^{(A)}(t-t')$ is referred to Eq. (10) for its definition. Similar as introduced in Eq. (25), we defined here $A^{(\sigma)^{\prime}}_{\alpha\mu\nu}(t) = \sum_{\nu'} \int_t^{t'} dt' C^{(\sigma)^{\prime}}_{\alpha\mu\nu}(t-t') \{ e^{-i\mathcal{L}(t-t')} [a_{\nu'} \bar{\rho}_j(t') \}]$.

For the convenience of application, we summarize the solving protocol in a more transparent way as follows. First, solve $\tilde{U}(\omega)$ from Eq. (9) and obtain $\rho(\omega)$ from Eq. (8); then, extract $\tilde{U}(\omega)$ from Eq. (28) and $N(\omega)$ from Eq. (27). With the help of $\tilde{U}(\omega)$, $\tilde{U}(\omega)$ and $N(\omega)$, one can straightforwardly calculate the noise spectrum of Eq. (26).

IV. ILLUSTRATIVE APPLICATIONS

A. Noninteracting Quantum Dot

We consider the simplest case of transport through a single-level quantum dot. In the absence of magnetic field and Coulomb interaction, the spin is an irrelevant degree of freedom which is thus neglected in this example. Then, the system Hamiltonian reads $H_S = \epsilon_0 a^\dagger a$, and the states involved in the transport are $|0\rangle$ and $|1\rangle$, corresponding to the empty and occupied dot states. Along the solving protocol outlined above, it is straightforward to obtain the shot noise spectrum as shown in Fig. 2 by the solid curve. Shown also there, by the dashed and dotted curves, are the results from the 2nd-order non-Markovian and Markovian master equation (2nd-nMKV/MKVM-E). The former is based on Ref. [18], while the later is from the following analytic result [19]:

$$S(\omega) = 2\bar{I} \left( \frac{\Gamma^2 + \Gamma_0^2 \epsilon^2 + \omega^2}{\Gamma^2 + \omega^2} \right), \tag{29}$$

where $\Gamma = \Gamma_L + \Gamma_R$ is assumed. $\bar{I}$ is the steady state current, in large bias limit which simply reads $\bar{I} = \Gamma_L \Gamma_R / \Gamma$, while here we account for the finite bias effect based on the SCBA-ME approach.
We observe that, quantitatively, the result from the $n$-SCBA-ME modifies that from the 2nd-nMKV-ME, while qualitatively both revealing a staircase behavior at frequency around $\omega_{a0} = |\mu_a - \epsilon_0|$. Mathematically, the origin of the staircase is from the time-nonlocal memory effect. Physically, this behavior is owing to the detection-energy $\langle \omega \rangle$ assisted transmission resonance between the dot and leads, which experiences a sharp change when crossing the Fermi levels. In high frequency regime, the noise spectrum from the $n$-SCBA-ME coincides with that from the 2nd-nMKV-ME, while the latter is given in Ref. [18] by the high frequency limit as $S(\omega \to \infty) = \Gamma_R$. This, straightforwardly, leads to a Fano factor as $F = S/2I = (1 + \Gamma_R/\Gamma_L)/2$. Therefore, it can be Poissonian, sub-Poissonian, and super-Poissonian, depending on the symmetry factor $\Gamma_R/\Gamma_L$. In contrast, the 2nd-nMKV-ME predicts a Poissonian result, $F(\omega \to \infty) = 1$.

We would like to remark that the 2nd-MKV-ME is only applicable in the low frequency regime of $\omega < \omega_{a0} = |\mu_a - \epsilon_0|$. This is in consistency with the fact that the high frequency regime corresponds to a short timescale where the non-Markovian effect is strong, while the low frequency regime corresponds to a long timescale where the non-Markovian effect diminishes.

\section*{B. Coulomb-Blockade Quantum Dot}

This is the system described by Eq. (18). Here we consider first the noise spectrum in the Coulomb-Blockade (CB) regime, while leaving the Kondo regime in next subsection. The CB regime of single occupation is characterized by $\epsilon_0 + U > \mu_L > \epsilon_0 > \mu_R$. For the purpose of comparison, we quote the result from the 2nd-MKV-ME

\begin{equation}
S(\omega) = 2I \left[ \frac{4\Gamma_L^2 + \Gamma_R^2 + \omega^2}{(2\Gamma_L + \Gamma_R)^2 + \omega^2} \right].
\end{equation}

In large bias limit, i.e., the Fermi levels being far from $\epsilon_0$ and $\epsilon_0 + U$, the steady state current reads $I = 2\Gamma_L \Gamma_R/(2\Gamma_L + \Gamma_R)$. However, in numerical simulation we account for the finite bias effect by inserting the steady state current from the SCBA-ME approach into Eq. (30). Notice that in obtaining Eq. (30) the double occupancy of the dot is excluded because its energy is out of the bias window. In the $n$-SCBA-ME treatment, however, all the four basis states should be included.

In Fig. 3 we display the main result of the noise spectrum in the CB regime, where a couple of non-Markovian resonance steps are revealed at frequencies around $\omega_{a0} = |\mu_a - \epsilon_0|$ and $\omega_{a1} = \epsilon_0 + U - \mu_a$. We find that the resonance steps in high frequency regime are enhanced by the Coulomb interaction, while the low frequency spectrum has remarkable “renormalization” effect compared to Eq. (30). In addition to the result under the wide band limit (WBL), in Fig. 3 we also show the bandwidth effect by two more curves. We see that, for finite-bandwidth leads, the noise spectrum diminishes at high frequency limit. This is because the energy $\langle \omega \rangle$ absorption/emission of detection restricts the channels for electron transfer between the dots and leads.

\section*{C. Nonequilibrium Kondo Dot}

The nonequilibrium Kondo system, with the Anderson impurity model realized by transport through a small
quantum dot, has been attracted intensive attention in the past two decades [20–30]. Compared to the equilibrium Kondo effect, the nonequilibrium is characterized by a finite chemical potential difference of the two leads. As a result, the peak of the density of states (spectral function) splits into two peaks pinned at each chemical potential. The two peak structure is difficult to probe directly, by the usual dc measurements. Nevertheless, the shot noise can be a promising quantity to reveal the nonequilibrium Kondo effect, although much less is known about it. We notice that results on low-frequency noise measurements have only appeared very recently [31, 32], while so far there are not yet reports on the finite-frequency (FF) noise measurements. A couple of theoretical studies [33–36], however, revealed diverse signatures (Kondo anomalies) in the FF noise spectra, such as an “upturn” [33] or a spectral “dip” [36] appeared at frequencies $\pm eV/\hbar$ ($V$ is the bias voltage), as well as the Kondo singularity (discontinuous slope) at frequencies $\pm 2eV/\hbar$ in Ref. [34], or at $\pm eV/2\hbar$ in Ref. [36]. Also, it was pointed out in Ref. [34] that the minimum (dip) developed at $\pm eV/\hbar$ is not relevant to the Kondo effect, since in the noninteracting case the noise has similar discontinuous slope at $\pm eV/\hbar$ as well.

The system Hamiltonian is still Eq. (18), which corresponds to the well known Anderson impurity model. Following the solving protocol outlined at the end of Sec. III together with the results in Sec. II (D), we obtain the noise spectrum in the Kondo regime as shown in Fig. 4. Remarkably, we notice a profound dip behavior (Kondo signature) in the noise spectrum at frequencies $\omega = \pm V/2$, as particularly demonstrated by a couple of voltages. We attribute this behavior to the emergence of the Kondo resonance levels (KRLs) at the Fermi surfaces, i.e., at $\mu_L = V/2$ and $\mu_R = -V/2$. In steady state transport, it is well known that the KRLs are clearly reflected in the spectral function. In terms of the master equation, the KRLs structure is hidden in the self-energy terms, which characterize the tunneling process and define the transport current. Similarly, the noise spectrum is essentially affected, particularly in the Kondo regime, by the self-energy process in frequency domain based on the same master equation. This explains the emergence of the spectral dip appearing at the same KRLs (i.e., at $\omega = \pm V/2$).

Alternatively, as a heuristic picture, one may imagine to include the KRLs as basis states in propagating $\rho(t)$, which is implied in the current correlation function. In usual case, when the level spacing is larger than its broadening, the diagonal elements of the density matrix decouple to the evolution of the off-diagonal elements. However, in the Kondo system, the diagonal and off-diagonal elements are coupled to each other, through the complicated self-energy processes. This feature would bring the coherence evolution described by the off-diagonal elements, with characteristic energies of the KRLs and their difference, into the diagonal elements which contribute directly to the second current measurement in the correlation function $\langle I(t)I(0) \rangle$. Then, one may expect three coherence energies, $\pm V/2$ and $V$, to participate in the noise spectrum. Indeed, the dip emerged in Fig. 4 reveals the coherence induced oscillation at the frequencies $\pm V/2$, while the other one at the higher frequency $V$ (observed in Ref. [36] in the case of infinite $U$) is smeared in our finite $U$ system by the rising noise with frequency.

V. SUMMARY

To summarize, in this work we propose a particle-number-resolved transport master equation under self-consistent Born approximation. The most advantage of this approach is its efficiency in the study of shot noise and a potential application in counting statistics. We have demonstrated this new approach by several examples, including particularly the nonequilibrium Kondo system. The obtained results are completely beyond the scope of the Born-Markov master equation, revealing such as staircase behavior and the profound nonequilibrium Kondo signature in the shot noise spectrum. The validity of the proposed approach is also supported by the evidence in steady state, where this approach can recover not only the exact result of noninteracting transport under arbitrary voltages, but also the challenging nonequilibrium Kondo effect.

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