Statistics of an adiabatic charge pump

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We investigate the effect of time-dependent cyclic-adiabatic driving on the charge transport in a quantum junction. We propose a nonequilibrium Green's function formalism to study the statistics of the charge pumped (at zero bias) through the junction. The formulation is used to demonstrate charge pumping in a single electronic level coupled to two (electronic) reservoirs with time-dependent couplings. An analytical expression for the average pumped current for a general cyclic driving is derived. It is found that for zero bias, for a certain class of driving, the Berry phase contributes only to the odd cumulants. In contrast, a quantum master equation formulation does not show a Berry-phase effect at all.

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I. INTRODUCTION

It is well known that the effect of adiabatically varying a few parameters in the Hamiltonian in a cyclic manner enters in the wave function in the form of a phase factor. This phase factor consists of two parts; one is called the dynamical part (which, in general, depends on how fast the parameters are varied) and the second one is generally known as the Berry phase (also called the geometric phase), which depends only on the path (area) traced in the parameter space and is independent of how fast it is traced provided the adiabatic condition is satisfied [1–3]. Somewhat counterintuitively, this phase factor may lead to changes in macroscopic observables, such as finite spin or charge currents in one-dimensional phase-coherent rings at equilibrium [4–6]. Originally developed in the context of closed quantum systems, recent works have extended the geometric phase concept to the case of open quantum systems out of equilibrium [7,8]. Adiabatic cyclic variation of parameters in the Hamiltonian leads to finite flux [9–11] in open systems. Switkes *et al.* [12] have experimentally demonstrated an adiabatic quantum pump by modulating the confining potential of an open quantum dot in a cyclic manner, leading to a finite voltage drop across the quantum dot. Modifying the potential at two ends of the dot changes the character of the wave function and therefore modifies the couplings to the electron reservoirs. In this work we explore this aspect within the most general framework based on the nonequilibrium Green's function [13]. The adiabatic driving may also affect the statistics of charge transfer, and the steady-state fluctuation relation due to Gallavoti-Cohen (GC) type symmetry [14] may also get modified. Ren *et al.* [11] have recently used quantum master equations (QMEs) to study heat pumping and fluctuations of heat transfer in a two-level system sandwiched between two thermal reservoirs. It was shown that in the case of time-dependent temperature modulations of the two heat reservoirs, heat transfer statistics do not admit GC-type symmetry. It was also argued that modulating couplings to thermal reservoirs does not lead to any pumping. Several methods such as scattering theory [15,16], Floquet scattering theory [17], and the adiabatic master equation approach [8,10] have been developed for studying the statistics of adiabatic pumping. But each of these methods has its own advantages and shortcomings. We note that in Ref. [18], the nonequilibrium Green's function (NEGF) method (which in principle is exact) has been used to study the pumped charge flux in interacting quantum dots. In this work we develop a scheme within the NEGF formalism to study statistics of the pumped charge transfer due to cyclic-adiabatic driving and apply it to a resonant level model. We present an analytic expression of the pumped charge for a general adiabatic-cyclic modulation of the level couplings with reservoirs. The direction of the net charge flow can be varied by changing the sign of the phase difference between the two time-dependent couplings. We find that the flux direction also depends on the energy difference, $\mu - \omega_0$, between the chemical potentials (μ) of reservoirs and the level energy (ω_0). In general, the charge transfer fluctuations are modified due to the Berry phase. However, at equilibrium, for a certain class of drivings, only asymmetric fluctuations (odd cumulants) are generated due to the Berry phase. The full statistics of the pumped charge satisfy a steady-state fluctuation relation. We emphasize that the present formulation shows that it is possible to pump a finite net charge in *noninteracting* open quantum junctions, unlike a simple QME formulation which does not lead to any pumping due to the Berry phase [10,11]. Although we use the term Berry phase, it is used in the sense of geometric contribution.

In Sec. II we introduce the model Hamiltonian to describe electron transport in molecular junctions, and give a brief sketch of the two-point measurement method and derive a generating function (GF) to study full the statistics of charge transfer due to adiabatic time modulation. In Sec. III, we express the GF in terms of the Green's function of the system and then present an approximate scheme to compute the Green's function analytically. In Sec. IV we apply this formulation to study the statistics of the pumped charge in a resonant level model. Conclusions are drawn in Sec. V.

II. HAMILTONIAN AND TWO-POINT MEASUREMENT SCHEME

A general Hamiltonian for the description of electron transport in a quantum junction where a molecular system is coupled to two (noninteracting) electronic reservoirs is

$$\hat{H}(t) = \sum_r \epsilon_r(t) d_r^\dagger d_r + H_{\rm int} + \sum_{\alpha k} \epsilon_{\alpha,k} c_{\alpha k}^\dagger c_{\alpha k} + \sum_{r,\alpha k} [g_{\alpha k,r}^*(t) c_{\alpha k}^\dagger d_r + g_{\alpha k,r}(t) d_r^\dagger c_{\alpha k}], \quad (1)$$

where $d_r^\dagger$ (d_r) stands for the electron creation (annihilation) operator in the rth system orbital while $c_{\alpha,k}^\dagger$ ($c_{\alpha,k}$) is for the electron creation (annihilation) operator on the left or right ($\alpha = L/R$) reservoir in the energy state $\epsilon_{\alpha,k}$. $H_{\rm int}$ is the Hamiltonian to account for all other possible interactions in the system, such as Coulomb and electron-phonon interactions. Here system lead couplings and/or single electron orbital energies can be periodically modulated (which can be experimentally realized by applying time-dependent gate voltages). In this work we consider the case when the driving time period is large compared to the internal relaxation time scales in the system such that the system at any time is at steady state with respect to reservoirs.

Under these conditions, we consider two simultaneous measurements of electron number in the left and the right reservoirs (as $[N_L,N_R]=0$, simultaneous measurement is quantum mechanically allowed) at time T_0 and $T > T_0$. The probability of change in the number of particles (n_α) in the left and the right reservoirs during the measurement time, $T - T_0$, can be computed using the generating function (GF) containing corresponding counting parameters λ_L and λ_R as

$$P(n_L,n_R,T-T_0) = \int_0^{2\pi}\frac{d\lambda_L}{2\pi}\int_0^{2\pi}\frac{d\lambda_R}{2\pi} \mathcal{Z}(\lambda_L,\lambda_R,T-T_0)e^{i(\lambda_L n_L+\lambda_R n_R)}. \quad (2)$$

Using the two-time quantum measurement formalism the generating function for particle number counting on both reservoirs can be written as [14]

$$\mathcal{Z}(\lambda,T-T_0) = \left\langle e^{-i[\lambda_L N_L(T)+\lambda_R N_R(T)]}e^{i[\lambda_L N_L(T_0)+\lambda_R N_R(T_0)]}\right\rangle_{\rho(T_0)}, \quad (3)$$

where $\rho(T_0)$ is the density matrix of the system+reservoirs at time T_0 and $N_L = \sum_k c_{Lk}^\dagger c_{Lk}$, $N_R = \sum_k c_{Rk}^\dagger c_{Rk}$ are the particle number operators corresponding to left and right reservoirs, respectively. Assuming $[\rho(T_0),N_L]=0$ and $[\rho(T_0),N_R]=0$ (or, more generally, system and reservoirs are decoupled at time T_0 and each are at equilibrium independently), Eq. (3) can be recast as

$$\begin{aligned}\mathcal{Z}(\lambda_L,\lambda_R,T-T_0) &= \left\langle \mathcal{U}_{(\frac{\lambda_L}{2},\frac{\lambda_R}{2})}(T_0,T)\mathcal{U}_{(-\frac{\lambda_L}{2},-\frac{\lambda_R}{2})}(T,T_0)\right\rangle_{\rho(T_0)} \\ &= \left\langle \mathcal{T}_c e^{-\frac{i}{\hbar}\int_c H_T^{\lambda(\tau)}(\tau)d\tau}\right\rangle_{\rho(T_0)}, \end{aligned} \quad (4)$$

where in the last line time-dependent $\lambda(\tau)$ has been defined on the Keldysh contour [19,20] (which goes from T_0 to T and back to T_0) [21] as $\lambda(\tau) = (-\frac{\lambda_L}{2}, -\frac{\lambda_R}{2})$ on the forward contour and $\lambda(\tau) = (\frac{\lambda_L}{2}, \frac{\lambda_R}{2})$ on the backward contour. $\mathcal{T}_c$ refers to the time ordering operator on the Keldysh contour. The evolution on the Keldysh contour is with respect to the $\lambda(\tau)$-dependent Hamiltonian (note that λ-dependent evolution is no longer unitary), and evolves the ket and the bra with different λ-dependent Hamiltonians [22], which can be obtained by replacing the last line in Eq. (1) with $\sum_{r,\alpha k}[g_{\alpha k,r}^*(\tau)e^{i\lambda_\alpha(\tau)}c_{\alpha k}^\dagger d_r + g_{\alpha k,r}(\tau)e^{-i\lambda_\alpha(\tau)}d_r^\dagger c_{\alpha k}]$. Hence the effect of measurement is reflected in the form of modified (λ-dependent) couplings to the reservoirs [23].

III. GENERATING FUNCTION IN TERMS OF THE NONEQUILIBRIUM GREEN'S FUNCTION AND AN APPROXIMATE SCHEME TO COMPUTE THE NONEQUILIBRIUM GREEN'S FUNCTION

Taking the λ_R derivative [21] of the logarithm of Eq. (4), we get

$$\begin{aligned}&\left[\frac{\partial \ln\mathcal{Z}(\lambda_L,\lambda_R,T-T_0)}{\partial(i\lambda_R)}\right] \\ &= \frac{1}{2}\sum_{k,r}\int_{T_0}^{T} dt_1\big[g_{Rk,r}^*(t_1)e^{-i\frac{\lambda_R}{2}}G_{r,Rk}^{++}(t_1,t_1) \\ &\quad - g_{Rk,r}(t_1)e^{i\frac{\lambda_R}{2}}G_{Rk,r}^{++}(t_1,t_1) + g_{Rk,r}^*(t_1)e^{i\frac{\lambda_R}{2}}G_{r,Rk}^{--}(t_1,t_1) \\ &\quad - g_{Rk,r}(t_1)e^{-i\frac{\lambda_R}{2}}G_{Rk,r}^{--}(t_1,t_1)\big], \end{aligned} \quad (5)$$

where $G_{Rk,r}^{++}(t,t')$, $G_{r,Rk}^{++}(t,t')$, $G_{Rk,r}^{--}(t,t')$, and $G_{r,Rk}^{--}(t,t')$ are appropriate real-time projections of mixed contour ordered Green's functions between system orbitals and reservoir states defined as $G_{Rk,r}^c(\tau,\tau') = -\frac{i}{\hbar}\langle\mathcal{T}_c c_{Rk}(\tau)d_r^\dagger(\tau')\rangle$ and $G_{r,Rk}^c(\tau,\tau') = -\frac{i}{\hbar}\langle\mathcal{T}_c d_r(\tau)c_{Rk}^\dagger(\tau')\rangle$. Here the $+$ $(-)$ index refers to the time variable located on the upper (lower) Keldysh contour [19]. Equation (5) can be recast in terms of the system Green's function matrix alone [20], which can be expressed in terms of Wigner-transformed quantities [13]. In the large measurement time limit, Eq. (5) can be recast as (see Appendix)

$$\begin{aligned}&\left[\frac{\partial \ln\mathcal{Z}(\lambda_L,\lambda_R,T-T_0)}{\partial(i\lambda_R)}\right] \\ &= \int_{T_0}^{T} dt\int_{-\infty}^{+\infty}\frac{d\omega}{2\pi} \\ &\quad\times \mathrm{Tr}[\breve{\Sigma}_R^{+-}(\omega,t)\breve{\mathbb{G}}^{-+}(\omega,t) - \breve{\mathbb{G}}^{+-}(\omega,t)\breve{\Sigma}_R^{-+}(\omega,t)]. \end{aligned} \quad (6)$$

Here the trace is over all the system orbitals and $\Sigma_R^{+-}(\omega,t)$ and $\Sigma_R^{-+}(\omega,t)$ are (Wigner-transformed) self-energy matrices (in system orbital space) arising due to interaction with the right reservoir. $G^{+-}(\omega,t)$ and $G^{-+}(\omega,t)$ are Wigner transforms of real-time projections of system contour ordered Green's function matrices with elements

$$G_{mn}^c(\tau,\tau') = -\frac{i}{\hbar}\langle\mathcal{T}_c d_m(\tau)d_n^\dagger(\tau')\rangle, \quad (7)$$

where $\langle\cdots\rangle$ is the average with respect to the density matrix evolving on the Keldysh contour as defined in Eq. (4). In Eq. (6) we have used the notation $\breve{A}^{-\eta} = -A^{-\eta}$ and $\breve{A}^{+\eta} = A^{+\eta}$, where $\eta = \pm$, to simplify the following expressions. The expression for $[\frac{\partial \ln\mathcal{Z}(\lambda_L,\lambda_R,T-T_0)}{\partial(i\lambda_L)}]$ can be obtained by replacing R with L in Eq. (6). Hence all that we have to do to get the final expression for the GF is to calculate the $\lambda(\tau)$-dependent Green's functions appearing in Eq. (6). This is done in the following.

The λ-dependent Green's function matrix defined on the Keldysh contour [with matrix elements in the system orbital space defined in Eq. (7)] satisfies the following equation of motion [20] (also known as the left-Dyson equation) on the Keldysh contour:

$$\int_c d\tau_1 \left[G_0^{-1}(\tau,\tau_1) - \Sigma^c(\tau,\tau_1)\right] G^c(\tau_1,\tau') = \delta^c(\tau,\tau'), \quad (8)$$

where $[i\hbar\frac{\partial}{\partial\tau} - H_0(\tau)]G_0(\tau,\tau') = \delta^c(\tau,\tau')$ and $\Sigma^c(\tau,\tau') = \Sigma^c_{\rm int}(\tau,\tau') + \Sigma^c_{\rm leads}(\tau,\tau')$ is the total self-energy due to $H_{\rm int}$ and system-reservoir coupling. $\Sigma^c_{\rm leads}(\tau,\tau')$ has matrix elements

$$\Sigma^c_{{\rm leads},rr'}(\tau,\tau') = \sum_\alpha \sum_{k,k'} g_{\alpha k,r}(\tau) e^{-i[\lambda_a(\tau)-\lambda_a(\tau')]} \times G^0_{\alpha k,\alpha k'}(\tau,\tau') g^*_{\alpha k',r'}(\tau'). \quad (9)$$

Here $G^0_{\alpha k,\alpha k'}(\tau,\tau')$ is the contour-ordered Green's functions of the free reservoir. An explicit expression for $\Sigma^c_{\rm int}$ depends on $H_{\rm int}$ which we keep general. The above equation can be projected onto the real times to obtain equations of motion for the Keldysh matrix $\breve{\mathbb{G}}(t,t')$. By employing the Wigner transformation [19,20] Eq. (8) can be recast as

$$\breve{\mathbb{G}}(\omega,t) = \breve{\mathbb{G}}_{ad}(\omega,t) + \breve{\mathbb{G}}_{ad}(\omega,t)\Bigg(\breve{\mathbb{G}}^{-1}_{ad}(\omega,t) \times \left\{1 - \exp\left[-\frac{i}{2}\left(\overleftarrow{\partial}_t \overrightarrow{\partial}_\omega - \overleftarrow{\partial}_\omega \overrightarrow{\partial}_t\right)\right]\right\}\breve{\mathbb{G}}(\omega,t)\Bigg), \quad (10)$$

where $\breve{\mathbb{G}}(\omega,t)$ is the system Green's function matrix. $\overleftarrow{\partial}_t$, $\overrightarrow{\partial}_t$ represent the classical time t derivative acting on the function to its left or to its right, respectively; similarly $\overleftarrow{\partial}_\omega$, $\overrightarrow{\partial}_\omega$ represent ω derivatives. The adiabatic contribution, $\breve{\mathbb{G}}_{ad}$, to the Green's function satisfies the matrix equation,

$$\breve{\mathbb{G}}_{ad}(\omega,t) = \left[\breve{\mathbb{G}}_0^{-1}(\omega,t) - \breve{\Sigma}(\omega,t)\right]^{-1}. \quad (11)$$

This is similar to the usual steady-state Green's function but the parameters are replaced with time-dependent parameters. Here $\breve{\Sigma}(\omega,t)$ and $\breve{\mathbb{G}}_0(\omega,t)$ are, respectively, the self-energy and the noninteracting system Green's function matrices. The second term in Eq. (10) represents a correction due to time variation of the parameters. Note that Eq. (10) is exact. We solve Eq. (10) to lowest order in the time derivative by iterating the equation for $\breve{\mathbb{G}}$ perturbatively in terms of time derivatives of $\breve{\mathbb{G}}_{ad}$ and retaining only terms linear in first derivative in (classical) time:

$$\breve{\mathbb{G}}(\omega,t) = \breve{\mathbb{G}}_{ad}(\omega,t) + \frac{i}{2}\breve{\mathbb{G}}_{ad}(\omega,t)\left\{\breve{\mathbb{G}}^{-1}_{ad}(\omega,t), \breve{\mathbb{G}}_{ad}(\omega,t)\right\}, \quad (12)$$

where $\{,\}$ stand for Poisson bracket in t and ω variables. We note that both the left- and the right-Dyson equations lead to the same approximate Eq. (12). We also note that Eq. (12) for the $\lambda = 0$ case preserves all the symmetries (as a consequence of the unitary evolution) of the Green's functions (see Appendix). Equations (11) and (12) can be solved to obtain the λ-dependent Green's functions which can be used to compute charge transfer statistics using Eq. (6). In the following, we apply this formalism to study the Berry effect on the charge transfer statistics in a resonant level model. The approximation in Eq. (12) allows us to analyze the pumped current analytically.

IV. ADIABATICALLY DRIVEN RESONANT LEVEL MODEL

Consider a single electronic site connected via time-dependent hopping to two electronic reservoirs. The Hamiltonian describing this model is the same as in Eq. (1) with $H_{\rm int}(t) = 0$ and only a single system orbital with time-independent energy $\epsilon = \omega_0$. We assume that only the amplitudes of the system-reservoir couplings are varied adiabatically. We put $\hbar = 1$ in this section. We compute the adiabatic Green's functions and the lowest order nonadiabatic corrections using Eqs. (11) and (12) (see Appendix). Using these Green's functions in Eq. (6), we obtain an expression for $\frac{1}{(T-T_0)}[\frac{\partial \ln \mathcal{Z}(\lambda_L,\lambda_R,T-T_0)}{\partial(i\lambda_R)}]$ (see Appendix). In order to emphasize the Berry-phase effect, we consider the two reservoirs at the same thermodynamic equilibrium ($\mu_L = \mu_R = \mu$, i.e., zero external bias and the same inverse temperatures, $\beta_L = \beta_R = \beta$). From n_L and n_R measurements, we can obtain the statistics of net charge, $n = (n_L - n_R)/2$, transferred between the two reservoirs and the change in charge, $N = n_L + n_R$, on the system. The transformation $(n_L,n_R) \leftrightarrow (n,N)$ leads to $P(N,n,T-T_0)$ (see Appendix).

Statistics of particle change on the system. Since $\tilde{P}(N,T-T_0) = \sum_n \tilde{P}(N,n,T-T_0)$, the GF, $\tilde{Z}(\Lambda,T-T_0)$, for $\tilde{P}(N,T-T_0)$ satisfies $\frac{1}{(T-T_0)}[\frac{\partial \ln \tilde{Z}(\Lambda,T-T_0)}{\partial(i\Lambda)}] = 0$; thus $\tilde{Z}(\Lambda,T-T_0) = 1$ and therefore $\tilde{P}(N,T-T_0) = \delta_{N0}$. This means that fluctuations in the electron change on the system die in the long-time limit. This is due to the finite dimensionality of the system Hilbert space.

Statistics of particles exchanged between reservoirs. The generating function $\tilde{Z}(\lambda,T-T_0)$ for the probability distribution of n electrons transferred from the right to the left reservoir, $\tilde{P}(n,T-T_0) = \sum_N \tilde{P}(N,n,T-T_0)$, is obtained as (see Appendix)

$$\frac{1}{(T-T_0)}\left[\frac{\partial \ln \mathcal{Z}(\lambda,T-T_0)}{\partial(i\lambda)}\right] = \frac{1}{T_p}\int_0^{T_p} dt \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi\,\Delta(\omega,t)} \{\,\Gamma_L(t)\Gamma_R(t) f(\omega)[1-f(\omega)](e^{i\lambda} - e^{-i\lambda})\}$$

$$+\frac{1}{T_p}\int_0^{T_p} dt \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi}\Bigg[\frac{[\dot{\Gamma}_L(t)+\dot{\Gamma}_R(t)]}{4[\Delta(\omega,t)]^2}(\omega-\omega_0)f'(\omega)\{\Gamma_R^2(t)-\Gamma_L^2(t)+\Gamma_L(t)\Gamma_R(t)[1-2f(\omega)][e^{i\lambda}-e^{-i\lambda}]\}$$
$$-\frac{\Gamma_L(t)+\Gamma_R(t)}{2[\Delta(\omega,t)]^2}[\Gamma_L(t)\dot{\Gamma}_R(t)-\Gamma_R\dot{\Gamma}_L(t)]f(\omega)[1-f(\omega)][e^{i\lambda}+e^{-i\lambda}-2]\{[0.5-f(\omega)]+(\omega-\omega_0)f'(\omega)\}\Bigg], \quad (13)$$

where $f'(\omega)=\frac{\partial f(\omega)}{\partial\omega}$, $\Gamma_\alpha(t)=2\pi|g_\alpha(t)|^2\rho$, $\dot{\Gamma}_\alpha(t)=\frac{\partial\Gamma_\alpha(t)}{\partial t}$, $\Delta(\omega,t)=(\omega-\omega_0)^2+(\frac{\Gamma_L(t)+\Gamma_R(t)}{2})^2+\Gamma_L(t)\Gamma_R(t)f(\omega)[1-f(\omega)][e^{i\lambda}+e^{-i\lambda}-2]$, and T_p is the time period of driving which is assumed to be much larger than the internal relaxation time of the system. Here it is assumed that the second measurement is carried out after $q=\frac{T-T_0}{T_p}$ number of cycles of driving (even if it is not the case the error is insignificant for large measurement time, for which the present formalism is developed). The first integral in Eq. (13) is the so-called dynamical contribution (similar to steady-state contribution with parameters replaced with time-dependent quantities with a time averaging). The second integral can be converted to parameter integral in (Γ_L,Γ_R) space and is independent of how fast the parameters are varied provided we are in the cyclic adiabatic limit; thus it represents a Berry contribution. When the two drivings are identical $\Gamma_L(t)=\Gamma_R(t)$, the Berry contribution vanishes identically, and the area traced in the parameter space (Γ_L,Γ_R) is zero. Equation (13) allows us to compute the full statistics of net particles transferred between the left and right reservoirs. For example, the expression for the average pumped charge is obtained by setting $\lambda=0$ in Eq. (13). The average number of electrons pumped per cycle is obtained as

$$N_{\rm pump}=\frac{\beta}{8\pi^2}\int_0^{T_p} dt\,\partial_t[\Gamma_L(t)-\Gamma_R(t)]\,{\rm Im}\,\Psi^{(1)}(Z), \quad (14)$$

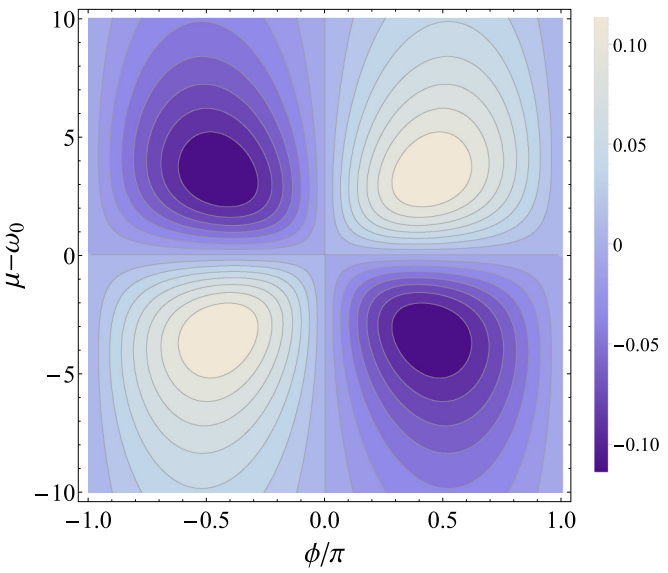

FIG. 1. Average number of particles pumped per cycle, $N_{\rm pump}$, as a function of $\mu-\omega_0$ and ϕ. Here $\Gamma_L(t)=1+10\sin^2(\frac{\pi t}{T_p}-\frac{\phi}{4})$, $\Gamma_R(t)=1+10\sin^2(\frac{\pi t}{T_p}+\frac{\phi}{4})$. Parameters Γ_α,μ, and ω_0 are in units of β^{-1}.

where $Z=\frac{1}{2}+i\frac{\beta}{2\pi}[\mu-\omega_0-i(\frac{\Gamma_L(t)+\Gamma_R(t)}{2})]$ and ${\rm Im}\,\Psi^{(1)}(Z)$ is the imaginary part of the trigamma function $\Psi^{(1)}(Z)$ of Z [24]. Note that Eq. (14) is valid for an arbitrary adiabatic driving, $\Gamma_\alpha(t)$. It is clear that $N_{\rm pump}=0$ when $\omega_0=\mu$ since ${\rm Im}\,\Psi^{(1)}(Z)=0$ in this case. This result may be useful in identifying resonance energy, ω_0, of an unknown quantum system at the junction by applying an external gate voltage on the system such that the net pumped charge is zero. Indeed, also when $\Gamma_L(t)=\Gamma_R(t)$, there is no net pumping of electrons between the two reservoirs. Thus the flux is purely driven due to Berry-phase effects. Additionally, the flux changes sign from $\mu>\omega_0$ to $\mu<\omega_0$ similarly to the case of small-amplitude driving [25]. This is demonstrated in Fig. 1 for sinusoidal drivings. Thus, both the phase difference between the drivings and the detuning act as driving forces that give rise to a net charge flux. Note that these forces are nonthermodynamic. The Berry phase changes sign as the driving is reversed (the area in parameter space is traced in reverse manner) and, as a consequence, the flux reverses the direction. Note that the dynamical part in Eq. (13) only contributes to the even cumulants. The Berry-phase part, on the other hand, in general, contributes to all the cumulants. However for even-cyclic drivings, i.e., $\Gamma_{L(R)}(-X)=\Gamma_{L(R)}(X)$ and $\Gamma_{L(R)}(t)\equiv\Gamma_{L(R)}(t/T_p\pm\phi/2)$, where ϕ is the phase difference, the Berry part contributes only to the odd cumulants. This fact may be helpful in designing experiments to distinguish the Berry contribution from the dynamical part. A plot of the first four cumulants as a function of phase difference between drivings ϕ is shown in Fig. 2. This figure demonstrate the periodicity of cumulants as a function of ϕ. It also shows that the magnitude of odd cumulants (purely due to the Berry

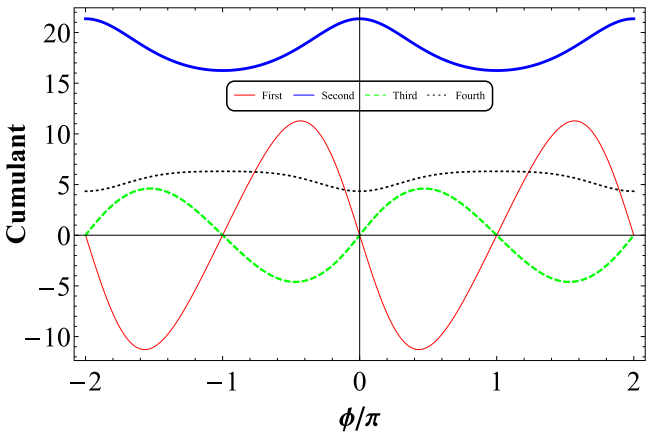

FIG. 2. Plot of first four cumulants ($\langle n\rangle$ and $\langle(n-\langle n\rangle)^k\rangle$ for $k=2,3,4$) per cycle as a function of ϕ. The drivings are the same as in Fig. 1. Here $\mu-\omega_0=3$ and $T_p=100$. Odd cumulants are magnified by 100 times for visual clarity.

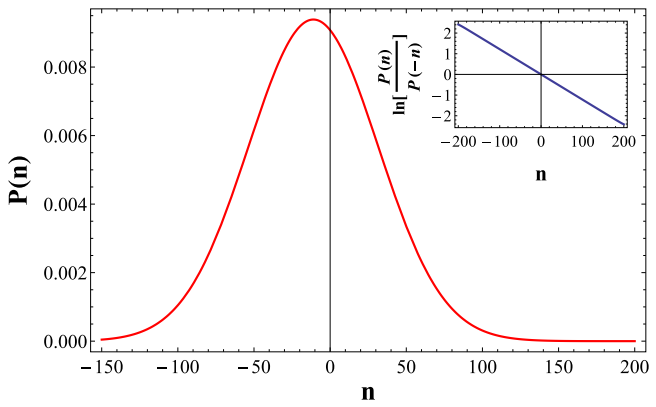

FIG. 3. Probability distribution for the net charge (n) transferred from the right to the left reservoir during the measurement time corresponding to $q = 100$ with $T_p = 100$, $\phi = \pi/2$, $\mu = 0$, and $\omega_0 = 3$. Time is in units of $h\beta$. Drivings are the same as in Fig. 1.

contribution) are order of $\frac{1}{T_p}$ smaller than the magnitude of the even cumulants. Further it is also clear from the figure that odd cumulants are odd functions of ϕ and even cumulants are even functions of ϕ.

In Fig. 3 we present numerical result for the full distribution function $P(n)$ for measurement time corresponding to $q = 100$. The distribution seems symmetric (second and third cumulants are $\sim$18 and $\sim$0.05, respectively) and is peaked around the value $n \sim qN_{\text{pump}}$. In the inset we show $\ln[P(n)/P(-n)]$ which grows linearly with n, confirming the GC-type fluctuation symmetry. However this symmetry originates due to nonthermodynamic forces and, as a consequence, the GFs for the forward ($\mathcal{Z}_F$) and backward ($\mathcal{Z}_B$) drivings satisfy $\mathcal{Z}_F(\lambda) = \mathcal{Z}_B(-\lambda)$ (see Appendix).

V. CONCLUSIONS

We developed an approximation scheme for computing the full counting statistics of adiabatic charge pumps within the NEGF formalism. We applied this formalism to study the statistics of pumped charge in a single-level model where the coupling to the reservoirs is driven adiabatically in time. It is found that a net (nonzero) number of electrons can be transferred between two reservoirs kept at the same thermodynamic states by adiabatically modulating the amplitudes of system-reservoir couplings. An analytic expression is derived for the net charge pumped per cycle entirely due to the geometric (Berry) phase. The phase difference between the drivings as well as the energy difference between the resonant level and reservoir chemical potential (at zero bias) are the important parameters that determine the direction of the pumped current. The statistics of the pumped charge is also influenced by the Berry phase, and the corresponding distribution function follows the Gollavati-Cohen type symmetry.

Note that in the present application we have assumed that only the amplitude of the system-reservoir couplings is varied adiabatically while the phase is constant. It is known that a simple linear time-dependent phase can be effectively transformed to the time-independent chemical potentials of the reservoirs [26]. Thus if phase is varied linearly with the same frequency, there is no net pumping of charge. For different frequency modulations, the net charge pumped per unit time is proportional to the difference of frequencies. This also can be obtained within the presented formulation in Sec. III. Consider system (resonant level)–reservoir couplings with a general time-dependent phase factor $|g_{\alpha k}|e^{i\phi_\alpha(t)}$ (where $\alpha = L/R$) such that the Hamiltonian is periodic in time with period T_p, assuming that the amplitude of $\phi_\alpha(t)$ is small so that adiabaticity remains valid at all times. Then, to first-order correction, Wigner-transformed self-energy due to couplings with the reservoirs in Eq. (11) is nonzero, unlike the case when only amplitudes are varied. This then leads to an expression for average pumped charge per cycle at zero bias as

$$N_{\text{pump}} = \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \frac{\Gamma_L \Gamma_R f'(\omega)}{(\omega-\omega_0)^2 + \left(\frac{\Gamma_L+\Gamma_R}{2}\right)^2} \times \int_0^{T_p} dt[\dot{\phi}_L(t) - \dot{\phi}_R(t)], \tag{15}$$

where $\Gamma_\alpha = 2\pi|g_\alpha|^2\rho$. This clearly demonstrates that if $\phi_\alpha(t)$ are considered as linear functions of t, i.e., $\phi_\alpha(t) = \omega_\alpha t + \phi$, there will be pumping only if $\omega_L \neq \omega_R$. Note that this result agrees with the exact result obtained by a time-dependent unitary transformation which transforms the linear phases in couplings to chemical potentials. On the other hand, if $\phi_\alpha(t)$ themselves are periodic functions in time, it is clear from Eq. (15) that the average pumped charge vanishes.

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APPENDIX

1. Derivation of Equation (6)

Equation (5) can be recast in terms of the system Green's function matrix alone by substituting the mixed Green's functions in terms of the system Green's functions [20]. We get

$$\left[\frac{\partial \ln \mathcal{Z}(\lambda_L,\lambda_R,T-T_0)}{\partial(i\lambda_R)}\right] = \int_{T_0}^{T} dt_1 \int_{T_0}^{T} dt_2 \text{Tr}[\Sigma_R^{+-}(t_1,t_2)G^{-+}(t_2,t_1) - G^{+-}(t_1,t_2)\Sigma_R^{-+}(t_2,t_1)]. \tag{A1}$$

Here the trace is over all the system orbitals and $\Sigma_R^{+-}(t,t')$, etc., are real-time projections of self-energy matrix $\Sigma_R(\tau,\tau')$ due to interaction with the right reservoir. It has matrix elements

$$\Sigma_{R;rr'}(\tau,\tau') = \sum_{k,k'} g_{Rk,r}(\tau)e^{-i[\lambda_R(\tau)-\lambda_R(\tau')]}G^0_{Rk,Rk'}(\tau,\tau')g^*_{Rk',r'}(\tau'), \tag{A2}$$

where $G^0_{Rk,Rk'}(\tau,\tau')$ are contour-ordered Green's functions of the free right reservoir. $G^{+-}(t,t')$, etc., are real-time projections of the system contour-ordered Green's function matrix with elements defined in Eq. (7). Now we use the Wigner representation of the quantities in the integrand of Eq. (A1) [19]; i.e., we use the transformation pair $A(t,t') = \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \mathbb{A}(\omega,t_c) e^{i\omega t_q}$ and $\mathbb{A}(\omega,t_c) = \int_{-\infty}^{+\infty} dt_q A(t,t') e^{-i\omega t_q}$ (here $t_c = \frac{t+t'}{2}$ and $t_q = t - t'$ are classical and quantum times, respectively) to get

$$\left[\frac{\partial \ln \mathcal{Z}(\lambda_L,\lambda_R,T-T_0)}{\partial(i\lambda_R)}\right] = \int_{T_0}^{T} dt_c \int_{-(T-T_0)}^{(T-T_0)} dt_q \int_{-\infty}^{+\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{+\infty} \frac{d\omega_2}{2\pi} e^{i(\omega_1-\omega_2)t_q} \times \mathrm{Tr}[\Sigma_R^{+-}(\omega_1,t_c)\mathbb{G}^{-+}(\omega_2,t_c) - \mathbb{G}^{+-}(\omega_1,t_c)\Sigma_R^{-+}(\omega_2,t_c)]. \tag{A3}$$

Here $t_c = \frac{t_1+t_2}{2}$ and $t_q = t_1 - t_2$. We neglect the effect of transients by assuming that the measurement time $T - T_0$ is large compared to internal relaxation times and the driving time period (hence we send the t_q integral from $-\infty$ to ∞). This leads to

$$\left[\frac{\partial \ln \mathcal{Z}(\lambda_L,\lambda_R,T-T_0)}{\partial(i\lambda_R)}\right] = \int_{T_0}^{T} dt \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \mathrm{Tr}[\Sigma_R^{+-}(\omega,t)\mathbb{G}^{-+}(\omega,t) - \mathbb{G}^{+-}(\omega,t)\Sigma_R^{-+}(\omega,t)]. \tag{A4}$$

2. Symmetrization of generating function

From $\mathcal{Z}(\lambda_L,\lambda_R,T-T_0)$ we can obtain the combined distribution function $P(n_L,n_R,T-T_0)$ for the electron number change on the left and right reservoirs over the measurement time period of $(T-T_0)$ using Eq. (2). However in order to compute the statistics of the net number (N) of electrons changed on both reservoirs (which is the same as the number of electrons changed on the system) and the net number (n) of electrons exchanged between both the reservoirs, we perform a coordinate transformation $(n_L,n_R) \to (N,n) = (\frac{n_L+n_R}{2}, \frac{n_L-n_R}{2})$. Performing this transformation in Eq. (2), we obtain

$$\tilde{P}(N,n,T-T_0) = \frac{1}{2}\int_0^{2\pi} \frac{d\Lambda}{2\pi} \int_{-2\pi}^{2\pi} \frac{d\lambda}{2\pi} \tilde{\mathcal{Z}}(\Lambda,\lambda,T-T_0) e^{i(\Lambda N+\lambda n)}, \tag{A5}$$

where the factor $1/2$ appears due to the Jacobian of transformation. $\tilde{\mathcal{Z}}(\Lambda,\lambda,T-T_0)$ is obtained from $\mathcal{Z}(\lambda_L,\lambda_R,T-T_0)$ by performing a coordinate transformation $(\lambda_L,\lambda_R) \to (\Lambda,\lambda) = (\lambda_L+\lambda_R,\lambda_L-\lambda_R)$. From $\tilde{P}(N,n,T-T_0)$ we can get $\tilde{P}(N,T-T_0)$ or $\tilde{P}(n,T-T_0)$ by summing over n or N, respectively. Summing over N, we get

$$\tilde{P}(n,T-T_0) = \frac{1}{2}\int_{-2\pi}^{2\pi} \frac{d\lambda}{2\pi} \tilde{\mathcal{Z}}(\lambda,T-T_0) e^{i\lambda n} = \int_0^{2\pi} \frac{d\lambda}{2\pi} \tilde{\mathcal{Z}}(\lambda,T-T_0) e^{i\lambda n}, \tag{A6}$$

where the second equality follows from the 2π periodicity of $\tilde{\mathcal{Z}}(\lambda,T-T_0)$.

3. λ-dependent Wigner-transformed Green's functions for the resonant level model

In this section we describe the procedure to get the λ-dependent Green's functions (up to lowest linear order correction in driving) for the resonant level model. We calculate the Green's functions on the Keldysh contour which goes from $-\infty$ to ∞ and back to ∞ under the large measurement time assumption in order to neglect initial correlations.

The noninteracting Green's function $\breve{\mathbb{G}}_0$ is defined below Eq. (8). The Wigner-transformed inverse of $\breve{\mathbb{G}}_0$ is given by

$$\breve{\mathbb{G}}_0^{-1}(\omega,t) = \begin{pmatrix} \omega - \omega_0 + i\eta & 0 \\ 0 & \omega - \omega_0 - i\eta \end{pmatrix},$$

where $\eta = 0^+$. The λ-dependent Wigner-transformed self-energy (with terms up to linear in first derivative in classical time) due to coupling to the reservoirs is $\breve{\Sigma}(\omega,t) = \sum_{\alpha=L,R} \breve{\Sigma}_\alpha(\omega,t)$ with

$$\breve{\Sigma}_\alpha(\omega,t) = \begin{pmatrix} -i\Gamma_\alpha(t)[0.5 - f_\alpha(\omega)] & i\Gamma_\alpha(t) f_\alpha(\omega) e^{i\lambda_\alpha} \\ i\Gamma_\alpha(t)[1 - f_\alpha(\omega)] e^{-i\lambda_\alpha} & i\Gamma_\alpha(t)[0.5 - f_\alpha(\omega)] \end{pmatrix}.$$

Here we assume that the system-reservoir couplings are real (the results remain same even for complex couplings with only the amplitudes being time dependent) and independent of energy (wide-band approximation). The first-order correction to the self-energy due to external driving is then zero (for the case when phases of couplings are time dependent, there will be a finite first order correction to self-energy).

Using Eq. (11) together with the above two equations for $\breve{\mathbb{G}}_0^{-1}$ and $\breve{\Sigma}_\alpha$, we get an expression for the adiabatic Green's function as

$$\breve{\mathbb{G}}_{ad}(\omega,t) = \frac{1}{\Delta(\omega,t)} \begin{pmatrix} \omega - \omega_0 - i\Gamma_L(t)[0.5 - f_L(\omega)] - i\Gamma_R(t)[0.5 - f_R(\omega)] & i\Gamma_L(t) f_L(\omega) e^{i\lambda_L} + i\Gamma_R(t) f_R(\omega) e^{i\lambda_R} \\ i\Gamma_L(t)[1 - f_L(\omega)] e^{-i\lambda_L} + i\Gamma_R(t)[1 - f_R(\omega)] e^{-i\lambda_R} & \omega - \omega_0 + i\Gamma_L(t)[0.5 - f_L(\omega)] + i\Gamma_R(t)[0.5 - f_R(\omega)] \end{pmatrix}$$

with

$$\Delta(\omega,t)=(\omega-\epsilon)^2+\left(\frac{\Gamma_L(t)+\Gamma_R(t)}{2}\right)^2+\Gamma_L(t)\Gamma_R(t)\{f_L(\omega)[1-f_R(\omega)](e^{i(\lambda_L-\lambda_R)}-1)+f_R(\omega)[1-f_L(\omega)](e^{-i(\lambda_L-\lambda_R)}-1)\},$$

where $f_L(\omega)$ and $f_R(\omega)$ are Fermi functions of the left and right reservoirs, respectively.

Using $\breve{\mathbb{G}}_{ad}(\omega,t)$ in Eq. (12) we calculate the lowest-order correction to the Green's functions. We give expressions only for $+-$ and $-+$ components:

$$\begin{aligned}\breve{\mathbb{G}}^{+-}(\omega,t)=&\frac{i\Gamma_L(t)f_L(\omega)e^{i\lambda_L}+i\Gamma_R(t)f_R(\omega)e^{i\lambda_R}}{\Delta(\omega,t)}-\frac{i[\dot\Gamma_L+\dot\Gamma_R](\omega-\omega_0)}{2[\Delta(\omega,t)]^2}[\Gamma_L f_L'(\omega)e^{i\lambda_L}+\Gamma_R f_R'(\omega)e^{i\lambda_R}]\\&+\frac{i[\Gamma_L(t)\dot\Gamma_R(t)-\Gamma_R(t)\dot\Gamma_L(t)]}{2[\Delta(\omega,t)]^2}\{[1-2f_R(\omega)]f_L(\omega)e^{i\lambda_L}-[1-2f_L(\omega)]f_R(\omega)e^{i\lambda_R}\}\\&+\frac{i(\omega-\omega_0)}{[\Delta(\omega,t)]^2}[\Gamma_L(t)\dot\Gamma_R(t)f_R(\omega)f_L'(\omega)-\Gamma_R(t)\dot\Gamma_L(t)f_L(\omega)f_R'(\omega)](e^{i\lambda_L}-e^{i\lambda_R}),\end{aligned}\tag{A7}$$

$$\begin{aligned}\breve{\mathbb{G}}^{-+}(\omega,t)=&\frac{i\Gamma_L(t)[1-f_L(\omega)]e^{-i\lambda_L}+i\Gamma_R(t)[1-f_R(\omega)]e^{-i\lambda_R}}{\Delta(\omega,t)}+\frac{i[\dot\Gamma_L+\dot\Gamma_R](\omega-\omega_0)}{2[\Delta(\omega,t)]^2}[\Gamma_L f_L'(\omega)e^{-i\lambda_L}+\Gamma_R f_R'(\omega)e^{-i\lambda_R}]\\&-\frac{i[\Gamma_L(t)\dot\Gamma_R(t)-\Gamma_R(t)\dot\Gamma_L(t)]}{2[\Delta(\omega,t)]^2}\{[1-2f_R(\omega)][1-f_L(\omega)]e^{-i\lambda_L}-[1-2f_L(\omega)][1-f_R(\omega)]e^{-i\lambda_R}\}\\&-\frac{i(\omega-\omega_0)}{[\Delta(\omega,t)]^2}\{\Gamma_L(t)\dot\Gamma_R(t)[1-f_R(\omega)]f_L'(\omega)-\Gamma_R(t)\dot\Gamma_L(t)[1-f_L(\omega)]f_R'(\omega)\}(e^{-i\lambda_L}-e^{-i\lambda_R}).\end{aligned}\tag{A8}$$

Using $\breve{\mathbb{G}}^{+-}(\omega,t)$, $\breve{\mathbb{G}}^{-+}(\omega,t)$, $\breve{\Sigma}_R^{+-}(\omega,t)$, and $\breve{\Sigma}_R^{-+}(\omega,t)$ in Eq. (6) we get an expression for $\frac{1}{(T-T_0)}[\frac{\partial\ln\mathcal{Z}(\lambda_L,\lambda_R,T-T_0)}{\partial(i\lambda_R)}]$ to lowest-order correction:

$$\begin{aligned}&\frac{1}{(T-T_0)}\left[\frac{\partial\ln\mathcal{Z}(\lambda_L,\lambda_R,T-T_0)}{\partial(i\lambda_R)}\right]\\&=\frac{1}{T_p}\int_0^{T_p}dt\int_{-\infty}^{+\infty}\frac{d\omega}{2\pi}\left\{\frac{\Gamma_L(t)\Gamma_R(t)}{\Delta(\omega,t)}\left\{f_R(\omega)[1-f_L(\omega)]e^{-i(\lambda_L-\lambda_R)}-f_L(\omega)[1-f_R(\omega)]e^{i(\lambda_L-\lambda_R)}\right\}\right.\\&+\left[\frac{\Gamma_R(t)[\Gamma_L(t)\dot\Gamma_R(t)-\Gamma_R(t)\dot\Gamma_L(t)]}{2[\Delta(\omega,t)]^2}[f_L(\omega)-f_R(\omega)]-\frac{\Gamma_R(t)[\dot\Gamma_L(t)+\dot\Gamma_R(t)](\omega-\omega_0)}{2[\Delta(\omega,t)]^2}[\Gamma_L(t)f_L'(\omega)+\Gamma_R(t)f_R'(\omega)]\right.\\&+\frac{\Gamma_R(t)[\Gamma_L(t)\dot\Gamma_R(t)-\Gamma_R(t)\dot\Gamma_L(t)]}{2[\Delta(\omega,t)]^2}[1-2f_R(\omega)]\left\{f_L(\omega)[1-f_R(\omega)](e^{i(\lambda_L-\lambda_R)}-1)+f_R(\omega)[1-f_L(\omega)](e^{-i(\lambda_L-\lambda_R)}-1)\right\}\\&-\frac{\Gamma_L(t)\Gamma_R(t)[\dot\Gamma_L(t)+\dot\Gamma_R(t)](\omega-\omega_0)}{2[\Delta(\omega,t)]^2}f_L'(\omega)\left\{[1-f_R(\omega)]\left(e^{i(\lambda_L-\lambda_R)}-1\right)+f_R(\omega)(e^{-i(\lambda_L-\lambda_R)}-1)\right\}\\&+\frac{\Gamma_L(t)\Gamma_R(t)}{[\Delta(\omega,t)]^2}\dot\Gamma_R(t)(\omega-\epsilon)f_L'(\omega)f_R(\omega)[1-f_R(\omega)]\left[e^{i(\lambda_L-\lambda_R)}+e^{-i(\lambda_L-\lambda_R)}-2\right]\\&\left.\left.-\frac{\Gamma_R^2(t)}{[\Delta(\omega,t)]^2}\dot\Gamma_L(t)f_R'(\omega)(\omega-\omega_0)\left\{f_L(\omega)[1-f_R(\omega)]\left(e^{i(\lambda_L-\lambda_R)}-1\right)+f_R(\omega)[1-f_L(\omega)]\left(e^{-i(\lambda_L-\lambda_R)}-1\right)\right\}\right]\right\},\end{aligned}\tag{A9}$$

where $f_\alpha'(\omega)$ is the ω derivative of $f_\alpha(\omega)$, $\dot\Gamma_\alpha(t)$ is the time derivative of $\Gamma_\alpha(t)$, and T_p is the time period of driving which is assumed to be much larger than the internal relaxation time of the system. A similar expression for $\frac{1}{(T-T_0)}[\frac{\partial\ln\mathcal{Z}(\lambda_L,\lambda_R,T-T_0)}{\partial(i\lambda_L)}]$ can be obtained by swapping L and R labels in Eq. (A9). Here it is assumed that the second measurement is carried out after an integer number of cycles $n=\frac{T-T_0}{T_p}$ of driving has been performed (even if it is not the case the error is minimal for large-time statistics, for which the present formalism is developed). Using expressions for $\frac{1}{(T-T_0)}[\frac{\partial\ln\mathcal{Z}(\lambda_L,\lambda_R,T-T_0)}{\partial(i\lambda_R)}]$ and $\frac{1}{(T-T_0)}[\frac{\partial\ln\mathcal{Z}(\lambda_L,\lambda_R,T-T_0)}{\partial(i\lambda_L)}]$, the expressions for the generating function for particle change on the system and particles exchanged between reservoirs can be obtained. These expressions for the $f_L(\omega)=f_R(\omega)=f(\omega)$ case are given in Sec. IV.

4. Symmetries of the Green's functions

As the full system is evolving with respect to a Hamiltonian, the evolution is unitary when $\lambda_L=\lambda_R=0$. As a consequence, matrix elements of $\breve{\mathbb{G}}(\tau,\tau')$ satisfy the symmetries [19]: (i) $[\breve{\mathbb{G}}_{mn}^{++}(t,t')]^*=\breve{\mathbb{G}}_{nm}^{--}(t',t)$, (ii) $[\breve{\mathbb{G}}_{mn}^{--}(t,t')]^*=\breve{\mathbb{G}}_{nm}^{++}(t',t)$, (iii) $[\breve{\mathbb{G}}_{mn}^{+-}(t,t')]^*=-\breve{\mathbb{G}}_{nm}^{+-}(t',t)$, and (iv) $[\breve{\mathbb{G}}_{mn}^{-+}(t,t')]^*=-\breve{\mathbb{G}}_{nm}^{-+}(t',t)$. These symmetries in the Wigner representation can be

summarized in a matrix form as

$$\breve{\mathbb{G}}(\omega,t)=\begin{bmatrix}\breve{\mathbb{G}}^{++}(\omega,t) & \breve{\mathbb{G}}^{+-}(\omega,t)\\ \breve{\mathbb{G}}^{-+}(\omega,t) & \breve{\mathbb{G}}^{--}(\omega,t)\end{bmatrix}\Longrightarrow[\breve{\mathbb{G}}(\omega,t)]^{*}=\begin{bmatrix}[\breve{\mathbb{G}}^{--}(\omega,t)]^{T} & -[\breve{\mathbb{G}}^{+-}(\omega,t)]^{T}\\ -[\breve{\mathbb{G}}^{-+}(\omega,t)]^{T} & [\breve{\mathbb{G}}^{++}(\omega,t)]^{T}\end{bmatrix}.$$

The approximate Green's function matrix used in this work can be expressed as

$$\breve{\mathbb{G}}(\omega,t)=\breve{\mathbb{G}}_{ad}(\omega,t)+\frac{i}{2}\left[\{\partial_{\omega}\breve{\mathbb{G}}_{ad}(\omega,t)\}\breve{\mathbb{G}}_{ad}^{-1}(\omega,t)\partial_{t}\breve{\mathbb{G}}_{ad}(\omega,t)-\{\partial_{t}\breve{\mathbb{G}}_{ad}(\omega,t)\}\breve{\mathbb{G}}_{ad}^{-1}(\omega,t)\partial_{\omega}\breve{\mathbb{G}}_{ad}(\omega,t)\right]. \tag{A10}$$

This approximate Green's function satisfies all the above symmetries provided $\breve{\mathbb{G}}_{ad}(\omega,t)$ satisfies the above symmetries which in turn requires $\breve{\mathbb{G}}_{0}(\omega,t)$ and $\breve{\Sigma}(\omega,t)$ to satisfy the above symmetries, which they do. Another important symmetry is $\breve{\mathbb{G}}_{mn}^{++}(t,t')-\breve{\mathbb{G}}_{mn}^{--}(t,t')=\breve{\mathbb{G}}_{mn}^{+-}(t,t')-\breve{\mathbb{G}}_{mn}^{-+}(t,t')$ which in the Wigner representation becomes $\breve{\mathbb{G}}_{mn}^{++}(\omega,t)-\breve{\mathbb{G}}_{mn}^{--}(\omega,t)=\breve{\mathbb{G}}_{mn}^{+-}(\omega,t)-\breve{\mathbb{G}}_{mn}^{-+}(\omega,t)$, which is also satisfied by the above approximate Green's function provided $\breve{\mathbb{G}}_{0}(\omega,t)$ and $\breve{\Sigma}(\omega,t)$ also satisfy it, which they do.

5. Detailed fluctuation theorem for pumped charge

We consider a special case when both the reservoirs are at the same thermodynamic states (i.e., $\beta_L=\beta_R=\beta$ and $\mu_L=\mu_R=\mu$) and drivings are of the form $\Gamma_L(t)=\Gamma(\frac{2\pi t}{T_p}-\frac{\phi}{2})$ and $\Gamma_R(t)=\Gamma(\frac{2\pi t}{T_p}+\frac{\phi}{2})$ [where $\Gamma(x)$ is an even periodic function with period 2π]. During the time-reversed evolution, the drivings at time t are obtained by substituting $t\to T_p-t$ for drivings in the forward evolution. Clearly the Hamiltonian does not have time-reversal symmetry as $\Gamma_{L(R)}(T_p-t)=\Gamma_{R(L)}(t)$; the left and right couplings switch roles. Then using Eq. (13), it can be shown that $\tilde{\mathcal{Z}}_F(\lambda,T-T_0)=\tilde{\mathcal{Z}}_B(-\lambda,T-T_0)$, where $\tilde{\mathcal{Z}}_F$ and $\tilde{\mathcal{Z}}_B$ are moment-generating functions of the probability distribution function for the number of particles exchanged between two reservoirs with forward and backward driving protocols, respectively. This is the Gallavotti-Cohen type symmetry for the zero-bias case. This symmetry leads to a detailed fluctuation theorem (FT)

$$\lim_{(T-T_0)\to\infty}\frac{P_F(n,T-T_0)}{P_B(-n,T-T_0)}=1, \tag{A11}$$

which is consistent with the standard (nondriven) steady-state FT for charge transfer in a single resonant level system at equilibrium (zero external bias). However, at steady state $P_F=P_B$, and the above relation leads to $P_F(n)=P_F(-n)$ at large measurement times. However, for a driven case, as we show in the Fig. 3, we find that $\tilde{\mathcal{Z}}_F(\lambda,T-T_0)\neq\tilde{\mathcal{Z}}_F(-\lambda,T-T_0)$ due to the Berry phase which leads to a finite net charge transfer between reservoirs.

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