# Energy transfer in the nonequilibrium spin-boson model: From weak to strong coupling

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To explore energy transfer in the nonequilibrium spin-boson model (NESB) from weak to strong system-bath coupling regimes, we propose a polaron-transformed nonequilibrium Green's function (NEGF) method. By combining the polaron transformation, we are able to treat the system-bath coupling nonperturbatively, thus in direct contrast to conventionally used NEGF methods which take the system-bath coupling as a perturbation. The Majorana-fermion representation is further utilized to evaluate terms in the Dyson series. This method not only allows us to deal with weak as well as strong coupling regimes but also enables an investigation on the role of bias in the energy transfer. As an application of the method, we study an Ohmic NESB. For an unbiased spin system, our energy current result smoothly bridges predictions of two benchmarks, namely, the quantum master equation and the nonequilibrium noninteracting blip approximation, a considerable improvement over existing theories. In case of a biased spin system, we found a bias-induced nonmonotonic behavior of the energy conductance in the intermediate coupling regime, resulting from the resonant character of the energy transfer. This finding may offer a nontrivial quantum control knob over energy transfer at the nanoscale.

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## I. INTRODUCTION

Over the last decades, energy transfer at the nanoscale has attracted significant attention and has grown considerably in importance. From an experimental perspective, the fabrication of nanoscale conductors such as various molecular junctions with desired properties can be achieved in the chemistry laboratory [1,2], meanwhile several procedures have been developed to measure energy transfer at a microscopic level, such as time-domain thermoreflectance techniques [3], scanning thermal microscopy [4], and laser Raman scattering thermometry [5,6]. Such experimental advances stimulated a surge of theoretical activity in understanding and controlling energy transfer in nanoscale conductors [7,8].

The nonequilibrium spin-boson model (NESB), as a minimum nontrivial molecular junction model, has its own advantages in studying energy transfer at nanoscale. On the one hand, the central two-level system has a diversity of physical realizations [9–12]. On the other hand, the total system is simple and tractable. As a result, its energy transfer characteristics are now an established area of theoretical research [13–15]. To date, various approaches have been carried out to deal with energy transfer in the NESB.

Theoretically, energy transfer processes in the NESB are usually investigated in two perturbative limits. When the system-bath coupling is weak, we can treat it in a perturbative manner. This treatment yields two eminent methods, namely, the quantum master equation (QME) analysis [13,16–18] and the nonequilibrium Green's function (NEGF) technique [19–21]. By adopting the Redfield approximation the QME can describe incoherent sequential transfer process [13,16] and it is even possible to include cotunneling process if the generalized Fermi golden rule is utilized [17]. Although simple and physically transparent analytical results are obtained, the QME is restricted to the weak coupling regime. The NEGF provides an alternative approach that formulates a formally exact expression for energy flux written in terms of the spin Green's functions (GFs). A perturbation expansion in the system-bath coupling is adopted only in obtaining those GFs. Velizhanin and coworkers [19] work out expressions for GFs using the Redfield approximation, however their results violate the energy conservation. In order to preserve conservation laws, a Majorana-fermion diagrammatic method [22–25] is further used in calculating GFs [21]. Compared with the QME, the NEGF can be applied in the intermediate coupling regime, but the strong coupling regime is still beyond its scope. In the opposite limit, when the system-bath coupling is strong, it is possible to choose the tunneling term as a perturbation. Along this line, we have the nonequilibrium version of the noninteracting blip approximation (NE-NIBA) [26,27] and a nonequilibrium polaron-transformed Redfield equation (NE-PTRE) method [28]. However, the NE-NIBA is valid only in the strong coupling regime [29]. The NE-PTRE, although it provides a correct picture for the NESB with super-Ohmic environments, reduces to the NE-NIBA framework for the NESB with Ohmic or sub-Ohmic ones [28] and thus faces the same pathology.

To go beyond perturbation theories, exact numerical techniques have been proposed, like multilayer multiconfiguration time-dependent Hartree theory [30], influence functional path integral techniques [31], and classical Monte Carlo simulations [32]. However, numerical simulations can become time consuming in the strong coupling and low temperature regimes, which limits their applicability.

So far, from both a theoretical and numerical point of view, a systematic investigation on the energy transfer process from weak to strong system-bath couplings for an unbiased spin system embedded in Ohmic bosonic baths is still absent [15]. Furthermore, little is known about the role that finite bias plays in the energy transfer of such systems. Only until very recently, the time-dependent energy transfer under the

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condition of a time-dependent bias has been studied within an influence functional approach [33]. However, the steady state properties still remain unknown. Note that the Ohmic bath serves as a paradigm for the simulation of environments with an abundance of low frequency modes, such as liquids, proteins, and polymers [34]; it is thus of utmost interest to introduce a systematic approach that allows for an arbitrary system-bath coupling and handles finite bias, to study energy transfer in NESB systems.

In this paper, we fill the gap and propose a unified theoretical scheme. Our strategy is to combine the polaron transformation (PT) [35] with the standard NEGF techniques, herein termed the polaron-transformed NEGF (PT-NEGF) method. The PT enables us to treat the system-bath coupling nonperturbatively [36,37], thus we can study the impact of system-bath coupling ranging from weak to strong regimes, in direct contrast to previous NEGF methods. Furthermore, in order to evaluate terms in the expansion series, we adopt the Majorana-fermion representation such that standard Feynman diagram techniques as well as the Dyson's equation can be applied to spin systems.

With the PT-NEGF, we investigate the energy transfer of the NESB embedded in the Ohmic bosonic baths in detail. For unbiased spin systems, our energy current result indeed smoothly bridges results of the QME and the NE-NIBA, two benchmarks which are valid in the weak and strong coupling regime, respectively. We also show that the method can cover a wide range of temperatures by comparing with an exact result at the Toulouse point [38] as well as with a noninteracting model, a fully harmonic thermal junction. In case of finite bias, we further found a nonmonotonic bias dependence of the conductance in an intermediate coupling regime with moderate temperatures. We attribute this dependence to the resonant character of the heat transfer. These features thus make our theory stand out from previous considerations.

The paper is organized as follows. We first introduce the NESB model and its nonequilibrium environment in Sec. II. In Sec. III, we present methodologies of the PT-NEGF. In Sec. IV, we study the energy transfer of the NESB using the PT-NEGF in detail. In Sec. V, we summarize our findings and make some final remarks.

## II. NONEQUILIBRIUM SPIN-BOSON SYSTEM

### A. Model

The NESB model, consisting of a two level system in contact with two bosonic reservoirs, is described by the general Hamiltonian [9]

$$H = H_s + H_I + H_B, \tag{1}$$

where the system Hamiltonian $H_s = \frac{\varepsilon}{2}\sigma_z + \frac{\Delta}{2}\sigma_x$ with $\varepsilon$ the bias, $\Delta$ the tunneling between two levels, and $\sigma_{x,z}$ the Pauli matrices. Since the spectrum of the system Hamiltonian is a symmetric function of bias, here we only consider positive bias. The bath part $H_B = \sum_{v=L,R} H_B^v = \sum_{j,v=L,R} \omega_{j,v} b_{j,v}^\dagger b_{j,v}$ and the bilinear interaction term $H_I = \sigma_z \sum_{j,v=L,R} g_{j,v}(b_{j,v}^\dagger + b_{j,v})$ with $b_{j,v}^\dagger (b_{j,v})$ the creation (annihilation) operator of the $j$th harmonic mode in the $v$ bosonic bath and $g_{j,v}$ the system-bath coupling strength. Throughout the paper, we set $\hbar = 1$ and $k_B = 1$. The influence of the bath is contained in the spectral density function:

$$I_v(\omega) = 2\pi \sum_{j\in v} g_{j,v}^2 \delta(\omega - \omega_{j,v}). \tag{2}$$

For convenience, we make the specific choice [38]

$$I_v(\omega) = \pi \alpha_v \omega^s \omega_c^{1-s} e^{-\omega/\omega_c}, \tag{3}$$

where $\alpha_v$ is the dimensionless system-bath coupling strength between the $v$ bath and the spin system, and $\omega_c$ is the cutoff frequency of the bath (we choose the same cutoff frequency for two baths). The case $s > 1 (s < 1)$ corresponds to super-Ohmic (sub-Ohmic) dissipation, and $s = 1$ represents the important case of frequency-independent (Ohmic) dissipation. In this paper, we limit ourselves to Ohmic dissipations.

### B. Energy current and energy conductance

We utilize the following definition for the energy current from the $v$th bosonic bath to the system:

$$J_v = -\frac{\partial}{\partial t}\langle H_B^v \rangle \tag{4}$$

with $H_B^v = \sum_j \omega_{j,v} b_{j,v}^\dagger b_{j,v}$ the Hamiltonian of the $v$th bosonic bath. It is worthwhile to mention that the above definition is consistent with the quantum thermodynamics and can be applied in the strong coupling regime [39].

We introduce GFs of Pauli matrices on the Keldysh contour [40,41]

$$\Pi_{\alpha\beta}(t,t') = -i\langle T_c \sigma_\alpha(t)\sigma_\beta(t')\rangle, \quad \alpha,\beta = x,y,z, \tag{5}$$

where $T_c$ is the contour-ordered operator responsible for the rearrangement of operators according to their contour time. The earlier (later) contour time places operators to the right (left). Its retarded, advanced, lesser, and greater components are given by $\Pi_{\alpha\beta}^r(t,t') = -i\Theta(t-t')\langle[\sigma_\alpha(t),\sigma_\beta(t')]\rangle$, $\Pi_{\alpha\beta}^a(t,t') = i\Theta(t'-t)\langle[\sigma_\alpha(t),\sigma_\beta(t')]\rangle$, $\Pi_{\alpha\beta}^<(t,t') = -i\langle\sigma_\beta(t')\sigma_\alpha(t)\rangle$, and $\Pi_{\alpha\beta}^>(t,t') = -i\langle\sigma_\alpha(t)\sigma_\beta(t')\rangle$, respectively, where $\Theta(\tau)$ denotes the Heaviside step function and $\sigma_\alpha(\tau) \equiv e^{iH\tau}\sigma_\alpha e^{-iH\tau}$ denotes the operator in the Heisenberg picture.

Noting that in nonequilibrium steady states the Keldysh GFs depend only on the time difference, $t - t'$. Therefore, in terms of Keldysh GFs defined above, a formally exact expression for the energy current from the $v$th bosonic bath to the system can be expressed as [21,32,42]

$$J_v = \frac{1}{2\pi}\int_0^\infty d\omega\omega I_v(\omega)[2n_v(\omega)\tilde{\chi}_z''(\omega) + \mathrm{Im}\Pi_{zz}^<(\omega)], \tag{6}$$

where $\Pi_{zz}^<(\omega)$ is the Fourier transform of the lesser Green's function $\Pi_{zz}^<(t-t')$, "Im" denotes the imaginary part, and $n_v$ is the Bose-Einstein distribution of temperature $T_v$. $\tilde{\chi}_z''(\omega)$ represents the imaginary part of the dynamical susceptibility; it is given by $-\mathrm{Im}\Pi_{zz}^r(\omega)$, or equivalently $[\Pi_{zz}^<(\omega) - \Pi_{zz}^>(\omega)]/2i$.

By considering zero dimensionality of the spin system and the conservation law of current $J_L + J_R = 0$, the above energy current formula [Eq. (6)] can be cast into a Landauer-type

form [32]:

$$J_L = \frac{\alpha\xi}{4\pi}\int_0^\infty d\omega\omega\tilde{\chi}_z''(\omega)\tilde{I}(\omega)[n_L(\omega)-n_R(\omega)], \quad (7)$$

where $\alpha=\alpha_L+\alpha_R$, and $\xi=4\alpha_L\alpha_R/\alpha^2$ is an asymmetry factor; for Ohmic dissipations, we have $\tilde{I}(\omega)=\pi\omega e^{-\omega/\omega_c}$. We should emphasize that Eq. (7) is still exact. In the linear response regime, the energy conductance defined by $\kappa\equiv dJ_L/dT_L|_{T_L\to T_R=T}$ is given by

$$\kappa = \frac{\alpha\xi}{4\pi}\int_0^\infty d\omega\,\tilde{\chi}_z''(\omega)\big|_{T_L=T_R=T}\tilde{I}(\omega)\left[\frac{\omega/2T}{\sinh(\omega/2T)}\right]^2. \quad (8)$$

From Eqs. (7) and (8), we can see that the imaginary part of the dynamical susceptibility $\tilde{\chi}_z''(\omega)$ determines the energy transfer properties of the NESB. Therefore, a unified description of $\tilde{\chi}_z''(\omega)$ results in a unified theory for energy transfer. In the following section, we will show that by developing a PT-NEGF method a quite general yet simple expression for $\tilde{\chi}_z''(\omega)$ can be obtained.

## III. POLARON TRANSFORMED NEGF METHOD

### A. Polaron transformation

Since $\tilde{\chi}_z''(\omega)$ is of primary interest, we will mainly focus on the calculation of the Keldysh GFs of $\sigma_z$ in the following, but the methodologies discussed below carry over easily to other Keldysh GFs as well [43]. We principally work here in the so-called nonadiabatic limit of $\Delta/\omega_c \ll 1$. For fast baths, it has been demonstrated that the PT is suitable for the entire range of system-bath coupling strength [35,37]. Thus we make the PT with the unitary operator

$$U=\exp[i\sigma_z\Omega/2], \quad \Omega=2i\sum_{j,v}\frac{g_{j,v}}{\omega_{j,v}}(b_{j,v}^\dagger-b_{j,v}) \quad (9)$$

on the Hamiltonian Eq. (1) such that

$$H_T=U^\dagger HU=\tilde{H}_0+\tilde{H}_I, \quad (10)$$

where the total free Hamiltonian is $\tilde{H}_0=\tilde{H}_s+\tilde{H}_B$ and the transformed system Hamiltonian reads

$$\tilde{H}_s=\frac{\varepsilon}{2}\sigma_z, \quad (11)$$

and the bath Hamiltonian remains unaffected, $\tilde{H}_B=H_B$. The transformed interaction term, originating from the tunneling term in Eq. (1), takes the following form:

$$\tilde{H}_I=\frac{\Delta}{2}(\sigma_x\cos\Omega+\sigma_y\sin\Omega). \quad (12)$$

It is evident that $\tilde{H}_I$ contains arbitrary orders of the system-bath coupling strength by noting the form of $\Omega$ in Eq. (9).

In order that a perturbation theory can be developed for $H_T$, $\langle\tilde{H}_I\rangle_{\tilde{H}_0}=0$ should be fulfilled. For the bath spectral function we choose [see Eq. (3)], it can be easily verified that for $s\leqslant 1$ the expectation of $\tilde{H}_I$ will always approach zero regardless of the system-bath coupling strength, while for $s>1$ the expectation is finite [28]. Therefore, in case of Ohmic dissipations we consider here, $\tilde{H}_I$ can be safely treated as a perturbation. The extension to the super-Ohmic dissipation is quite straightforward, and we should adopt a fluctuation-decoupling scheme [28,35] and choose $\tilde{H}_I-\langle\tilde{H}_I\rangle_{\tilde{H}_0}$ as the transformed interaction term. Noting $\tilde{H}_I$ is nonperturbative in coupling strength $\alpha$, we thus follow a totally different routine to develop a NEGF method.

Furthermore, since $[\sigma_z,U]=0$, expressions for the energy current as well as the heat conductance [see Eqs. (7) and (8)] are unchanged after the PT. Therefore, we still only need to evaluate the Keldysh GFs of $\sigma_z$ with respect to the transformed Hamiltonian $H_T$.

### B. Majorana-fermion representation

Note that spin operators do not satisfy the Wick theorem. In order to overcome this difficulty, the so-called Majorana-fermion representation (MFR) [44] is utilized in the method such that standard Feynman diagram techniques as well as the Dyson equation can be used. Technically, the MFR involves the introduction of a triplet of real fermions $\eta_\alpha$ (with $\alpha=x,y,z$) that satisfy [23]

$$\eta_\alpha\eta_\beta=-\eta_\beta\eta_\alpha\ (\alpha\neq\beta), \quad \eta_\alpha^2=1, \quad (13)$$

which leads to a representation of spin operators:

$$\sigma_\alpha=-i\sum_{\beta\gamma}\epsilon_{\alpha\beta\gamma}\eta_\beta\eta_\gamma. \quad (14)$$

Noting the crucial property of the MFR $\langle\sigma_\alpha(\tau)\sigma_\beta\rangle=\langle\eta_\alpha(\tau)\eta_\beta\rangle$ [22,23,25], if we introduce the Keldysh GFs of Majorana fermions

$$G_{\alpha\beta}(t,t')\equiv-i\langle T_c\eta_\alpha(t)\eta_\beta(t')\rangle, \quad (15)$$

then Keldysh GFs of spin operators can be rewritten in terms of the greater and lesser Keldysh GF of Majorana fermions, namely, $\Pi_{\alpha\beta}^<(t,t')=-G_{\alpha\beta}^<(t,t')$, $\Pi_{\alpha\beta}^>(t,t')=G_{\alpha\beta}^>(t,t')$, $\Pi_{\alpha\beta}^r(t,t')=\Theta(t-t')[G_{\alpha\beta}^>(t,t')+G_{\alpha\beta}^<(t,t')]$, $\Pi_{\alpha\beta}^a(t,t')=-\Theta(t'-t)[G_{\alpha\beta}^>(t,t')+G_{\alpha\beta}^<(t,t')]$. Thus the evaluation of Keldysh GFs $\Pi_{\alpha\beta}$ turns into an evaluation of Keldysh GFs $G_{\alpha\beta}$. The latter enables a standard diagrammatic method. For later convenience, we denote $G_\eta\equiv G_{zz}$, $G_{\eta_x}\equiv G_{xx}$, $G_{\eta_y}\equiv G_{yy}$.

### C. Evaluation of Keldysh Green's functions

By introducing the contour ordering, Keldysh GFs are formally and structurally equivalent to equilibrium counterparts [40,41]. So the Keldysh GF $G_\eta(t,t')$ satisfies a Dyson-like equation:

$$G_\eta(t,t')=G_{\eta,0}(t,t')+\int dt_1\int dt_2 G_{\eta,0}(t,t_1)\times\Sigma_\eta(t_1,t_2)G_\eta(t_2,t'), \quad (16)$$

where $G_{\eta,0}(t,t')$ is the free Keldysh GF of Majorana fermions, and $\Sigma_\eta(t_1,t_2)$ corresponds to the self-energy due to the system-bath interaction. Once the information on $G_{\eta,0}(t,t')$ and $\Sigma_\eta(t_1,t_2)$ is known, we can obtain $G_\eta(t,t')$ via the above equation.

#### 1. Free Keldysh Green's functions

We first consider free Keldysh GFs for Majorana fermions. In the MFR, the transformed system Hamiltonian $H_s$ can be

expressed as

$$\tilde{H}_s = -i\frac{\varepsilon}{2}\eta_x\eta_y. \tag{17}$$

Clearly we have $[\tilde{H}_s,\eta_z]=0$, so $\eta_z$ is time independent. We find in steady states ($\tau = t-t'$) that

$$G^{r/a}_{\eta,0}(\tau) = \mp 2i\Theta(\pm\tau), \tag{18}$$

which yields

$$G^{r/a}_{\eta,0}(\omega) = \frac{2}{\omega \pm i\xi} \tag{19}$$

with $\xi \to 0^+$ in the frequency domain.

We also need greater and lesser components of $G_{\eta_x,0}$ and $G_{\eta_y,0}$ in the calculations for self-energies below. From the Hamiltonian [Eq. (17)], equations of motion that $\eta_x$ and $\eta_y$ satisfy are given by $\eta_y(t) = \eta_y\cos\varepsilon t + \eta_x\sin\varepsilon t$ and $\eta_x(t) = \eta_x\cos\varepsilon t - \eta_y\sin\varepsilon t$, respectively. With a nonzero bias, the spin-down and spin-up states of $\sigma_z$ are no longer degenerate; for self-consistence and convenience, we choose the ground state, namely, the spin-down state (since $\varepsilon > 0$ by default) as the initial condition such that

$$\begin{aligned} G^{>}_{\eta_x,0}(\omega) &= G^{>}_{\eta_y,0}(\omega) = -i2\pi\delta(\omega-\varepsilon),\\ G^{<}_{\eta_x,0}(\omega) &= G^{<}_{\eta_y,0}(\omega) = i2\pi\delta(\omega+\varepsilon). \end{aligned} \tag{20}$$

It is worthwhile to mention that if we only consider negative values of bias (due to the symmetry, we can consider either positive or negative bias), we should take the spin-up state (the corresponding ground state) as the initial state, but the resulting final expressions remain the same forms with those obtained below, implying that our results are symmetric functions of the bias as expected. The independence of our results on the initial conditions should also be understood in this manner.

We then turn to free Keldysh GFs for bath operators. According to Eq. (12), we introduce compact notations $B \equiv (\cos\Omega, \sin\Omega)^T$ and $B^\dagger \equiv (\cos\Omega, \sin\Omega)$ and define a matrix Keldysh GF for steady states:

$$G_{B,0}(\tau) \equiv -i\langle T_c B(\tau)B^\dagger\rangle. \tag{21}$$

Since the retarded and advanced components are totally determined by the lesser and greater ones, we only need $G^{>}_{B,0}(\tau) = -i\langle B(\tau)B^\dagger\rangle$ and $G^{<}_{B,0}(\tau) = -i\langle B^\dagger B(\tau)\rangle$. In order to simplify the calculation of matrix elements, we further introduce correlation functions $\Phi_{nm}(\tau) \equiv \frac{\Delta^2}{4}\langle e^{ni\Omega(\tau)}e^{mi\Omega}\rangle$ and $\tilde{\Phi}_{nm}(\tau) \equiv \frac{\Delta^2}{4}\langle e^{ni\Omega}e^{mi\Omega(\tau)}\rangle = \Phi_{nm}(-\tau)$ with $n,m=\pm$. In terms of those correlation functions, we can rewrite matrix elements of $G^{>,<}_{B,0}$; for instance, we have

$$\begin{aligned} \langle\cos\Omega(\tau)\cos\Omega\rangle = \frac{1}{\Delta^2}[&\Phi_{++}(\tau)+\Phi_{+-}(\tau)\\ &+\Phi_{-+}(\tau)+\Phi_{--}(\tau)] \end{aligned} \tag{22}$$

for an element of $G^{>}_{B,0}$. By replacing $\Phi_{nm}(\tau)$ with $\tilde{\Phi}_{nm}(\tau)$, we can obtain results for elements of $G^{<}_{B,0}$.

By assuming the two reservoirs are at their own thermal equilibrium states characterized by temperature $T_v$ ($v=L,R$), $\Phi_{nm}(\tau)$ and $\tilde{\Phi}_{nm}(\tau)$ can be evaluated by using the techniques of Feynman disentangling of operators [36]. For example, we have [12,38]

$$\Phi_{+-}(\tau) = \frac{\Delta^2}{4}\exp\left[-\sum_{v=L,R}\left[Q^v_2(\tau)+iQ^v_1(\tau)\right]\right] \tag{23}$$

with $Q^v_1(\tau) = \frac{2}{\pi}\int_0^\infty d\omega\frac{I_v(\omega)}{\omega^2}\sin\omega\tau$ and $Q^v_2(\tau) = \frac{4}{\pi}\int_0^\infty d\omega\frac{I_v(\omega)}{\omega^2}\coth(\frac{\omega}{2T_v})\sin^2(\frac{\omega\tau}{2})$. The additive forms in exponential functions can be understood by noting the additive form of $\Omega$ for two baths [see Eq. (9)]. Similarly, we can find $\tilde{\Phi}_{+-}(\tau) = \tilde{\Phi}_{-+}(\tau) = \Phi^*_{+-}(\tau)$, $\Phi_{++}(\tau) = \Phi_{--}(\tau)$, and $\tilde{\Phi}_{++}(\tau) = \tilde{\Phi}_{--}(\tau)$. By noting, for instance, $\langle\cos\Omega(\tau)\sin\Omega\rangle = \frac{i}{\Delta^2}[\Phi_{+-}(\tau)-\Phi_{++}(\tau)+\Phi_{--}(\tau)-\Phi_{-+}(\tau)]$, we deduce that the nondiagonal elements of $G^{>,<}_{B,0}$ are vanishing, so $G^{>,<}_{B,0}$ are actually diagonal matrices.

### 2. *Self-energy*

We now focus on the extraction of the fermionic self-energies. In the MFR, the interaction part can be rewritten as

$$\tilde{H}_I \equiv -i\frac{\Delta}{2}(\eta_y\eta_z\cos\Omega + \eta_z\eta_x\sin\Omega). \tag{24}$$

For such a weak interaction, the spin dynamics should, in principle, be well described by the lowest-order self-energies. The lowest nonvanishing correction to $G_{\eta,0}$ is

$$\frac{i}{2}\int dt_1\int dt_2 \mathrm{Tr}\{\rho_0 T_c[\eta_z(t)\eta_z(t')\tilde{H}_I(t_1)\tilde{H}_I(t_2)]\} \tag{25}$$

with $\rho_0$ as the initial total density matrix. In what follows, we choose a factorized initial condition, namely, $\rho_0 = \rho_B\otimes\rho_s$. Both reservoirs are prepared in canonical-equilibrium states with temperatures $T_v \equiv \beta_v^{-1}$:

$$\rho_B = \prod_{v=L,R}\frac{e^{-\beta_v H^v_B}}{Z^v_B}, \quad Z^v_B = \mathrm{Tr}e^{-\beta_v H^v_B}. \tag{26}$$

The spin system $\tilde{H}_s$ is prepared in a spin-down state. From the above correction form, the leading order self-energy $\Sigma_\eta$ is obtained with both fermionic and bosonic lines given by the free propagators

$$\begin{aligned} \Sigma_\eta(t_1,t_2) = i\frac{\Delta^2}{4}[&{}_{11}G_{B,0}(t_1,t_2)G_{\eta_y,0}(t_1,t_2)\\ &+{}_{22}G_{B,0}(t_1,t_2)G_{\eta_x,0}(t_1,t_2)] \end{aligned} \tag{27}$$

with ${}_{11}G_{B,0} \equiv (1\quad 0)G_{B,0}\binom{1}{0}$ and ${}_{22}G_{B,0} \equiv (0\quad 1)G_{B,0}\binom{0}{1}$.

Using the Langreth theorem [41] and expressions for the free Keldysh GFs, we find the greater and lesser components of the self-energy $\Sigma_\eta$ can be written as $\Sigma^{>}_\eta(\omega) = -i\Phi_{+-}(\omega-\varepsilon)$ and $\Sigma^{<}_\eta(\omega) = i\tilde{\Phi}_{+-}(\omega+\varepsilon)$, respectively. Noting the property $\Sigma^r_\eta - \Sigma^a_\eta = \Sigma^{>}_\eta - \Sigma^{<}_\eta$, we have

$$\mathrm{Im}\left[\Sigma^{r/a}_\eta(\omega)\right] \equiv \mp\tfrac{1}{2}\Gamma(\omega) \tag{28}$$

with $\Gamma(\omega) \equiv [\Phi_{+-}(\omega-\varepsilon)+\tilde{\Phi}_{+-}(\omega+\varepsilon)]$. In order to capture correct physics in the strong coupling regime, the real part $\Lambda$ of self-energies $\Sigma^{r/a}_\eta(\omega)$, usually ignored in previous perturbation theory, should also be taken into account. Together

with Eq. (27) and Langreth theorems, we have

$$2\Lambda = \Sigma_\eta^r(\omega) + \Sigma_\eta^a(\omega) = i\frac{\Delta^2}{4}\Bigg[\sum_{\alpha=11,22}\left({}_\alpha G^r_{B,0} +_\alpha G^a_{B,0}\right)\circ\left(i\mathrm{Im}G^r_{\eta_y,0} + G^<_{\eta_y,0}\right) + \sum_{\alpha=11,22}({}_\alpha G^>_{B,0} +_\alpha G^<_{B,0})\circ \mathrm{Re}G^r_{\eta_y,0}\Bigg], \tag{29}$$

where "Im" and "Re" denote the imaginary and real part, respectively, and "$\circ$" denotes the convolution in the frequency domain. Introducing the Laplace transform for an arbitrary function $A(\tau)$,

$$A(\lambda) = \int_0^\infty A(\tau)e^{-\lambda\tau}d\tau, \tag{30}$$

and noting the Kramers-Kronig relations, we finally get

$$\Lambda(\omega) = \mathrm{Im}[\tilde{\Phi}_{+-}(\lambda)]|_{\lambda=-i(\omega+\varepsilon)} + \mathrm{Im}\,[\Phi_{+-}(\lambda)]|_{\lambda=-i(\omega-\varepsilon)}, \tag{31}$$

where $\Phi_{+-}(\lambda)$ and $\tilde{\Phi}_{+-}(\lambda)$ are bath correlations in the Laplace space. The sum of the greater and lesser self-energies gives the Keldysh component of self-energy:

$$\Sigma^K_\eta(\omega) = i[\tilde{\Phi}_{+-}(\omega+\varepsilon) - \Phi_{+-}(\omega-\varepsilon)]. \tag{32}$$

### D. Expression for $\tilde{\chi}''_z(\omega)$

Inserting free Keldysh GFs $G^{r/a}_{\eta,0}(\omega)$ and self-energies $\Sigma^{r/a}_\eta(\omega)$ into Eq. (16), we have

$$G^{r/a}_\eta(\omega) = \frac{2}{\omega - 2\Lambda \pm i\Gamma}. \tag{33}$$

The above results together with Eq. (32) lead to the following Keldysh component of $G_\eta$:

$$G^K_\eta(\omega) = \frac{4i[\tilde{\Phi}_{+-}(\omega+\varepsilon) - \Phi_{+-}(\omega-\varepsilon)]}{(\omega-2\Lambda)^2+\Gamma^2}. \tag{34}$$

Since $\tilde{\chi}''_z(\omega) = [\Pi^<_{zz}(\omega) - \Pi^>_{zz}(\omega)]/2i$, we have

$$\tilde{\chi}''_z(\omega) = \frac{i}{2}G^K_\eta = 2\frac{\Phi_{+-}(\omega-\varepsilon) - \tilde{\Phi}_{+-}(\omega+\varepsilon)}{(\omega-2\Lambda)^2+\Gamma^2}. \tag{35}$$

The above expression is one of the main results of this paper.

## IV. RESULTS AND DISCUSSION

In this section we shall apply the formal results of Eq. (35) to study the energy transfer for the case of Ohmic dissipation, that is, $s = 1$ in the spectral density Eq. (3).

### A. Unbiased system

We first consider unbiased spin systems with $\varepsilon = 0$. In this situation, Eq. (35) reduces to

$$\tilde{\chi}''_z(\omega) = 2\frac{\Phi_{+-}(\omega) - \tilde{\Phi}_{+-}(\omega)}{(\omega-2\Lambda)^2+\Gamma^2} \tag{36}$$

with $2\Lambda$ and $\Gamma$ now the imaginary and real part of $2[\Phi_{+-}(\lambda) + \tilde{\Phi}_{+-}(\lambda)]|_{\lambda=-i\omega}$, respectively. For later convenience, we introduce the correlation function of the $v$th bosonic bath:

$$C_v(\tau) = \frac{\Delta}{2}\exp\left[-Q^v_2(\tau) - iQ^v_1(\tau)\right], \tag{37}$$

therefore the bath correlation functions can be expressed as

$$\Phi_{+-}(\tau) = C_L(\tau)C_R(\tau), \quad \tilde{\Phi}_{+-}(\tau) = C_L(-\tau)C_R(-\tau), \tag{38}$$

and clearly the two baths are involved nonadditively.

In the nonadiabatic limit of $\omega_c/T_v \gg 1$, Eq. (37) has the following explicit form [12,38,45,46]:

$$C_v(\tau) = \frac{\Delta}{2}\exp[-i\pi\alpha_v\,\mathrm{sgn}(\tau)]\left(\frac{\pi T_v}{\omega_c\sinh(\pi T_v|\tau|)}\right)^{2\alpha_v}. \tag{39}$$

Taking the weak coupling limit of the above expression, after a straightforward calculation we find

$$\tilde{\chi}''_z(\omega) \simeq 2\frac{\Delta^2\sum_v I_v(\omega)}{(\omega^2-\Delta^2)^2 + \omega^2\left(\sum_v I_v(\omega)\coth\frac{\omega}{2T_v}\right)^2}. \tag{40}$$

Inserting the above expression into Eq. (7) yields

$$J_L = \frac{2}{\pi}\int_0^\infty d\omega\omega\frac{I_L(\omega)I_R(\omega)\Delta^2[n_L(\omega)-n_R(\omega)]}{(\omega^2-\Delta^2)^2 + \omega^2\left(\sum_v I_v(\omega)\coth\frac{\omega}{2T_v}\right)^2}, \tag{41}$$

which is exactly the result obtained by a previous NEGF method [21]. When the bath temperature $T_{L/R}$ is comparable to or larger than the energy spacing $\Delta$ of the spin, the incoherent sequential process becomes the dominant heat transfer mechanism [17], thus the integrand of Eq. (41) with frequencies around $\Delta$ contributes the most to the heat current. As a result, Eq. (41) can be reduced to the result of QME [21]:

$$J_L = \Delta\frac{I_L(\Delta)I_R(\Delta)[n_L(\Delta)-n_R(\Delta)]}{I_L(\Delta)[2n_L(\Delta)+1] + I_R(\Delta)[2n_R(\Delta)+1]}. \tag{42}$$

Thus our energy current formula can describe the weak coupling regime, in contrast to the NE-NIBA, the result [26,27]

$$J_L = \frac{1}{4\pi}\int_{-\infty}^\infty \omega[C_R(\omega)C_L(-\omega) - C_R(-\omega)C_L(\omega)]d\omega \tag{43}$$

of which can only be applied to the strong coupling regime [29].

In order to see the performance of our result in a wide range of the coupling strength, a comparison between various theoretical predictions for the energy current is shown in Fig. 1. From the figure, it is expected that our formula matches the results of the QME and the NEGF in the weak coupling regime, while the NE-NIBA underestimates values of the heat current. As the coupling strength increases, our result depicts a "turnover" behavior which is in accord with exact numerical results [30] as well as the NE-NIBA's prediction, thus our result is distinct from results of the QME and NEGF. This turnover phenomenon results from a renormalization effect of tunneling between two spin states in the strong coupling regime. In the strong coupling regime, the profile of our formula almost coincides with the result of the NE-NIBA. We attribute this agreement to the fact that approximations underlying our theory bear a close resemblance to that of

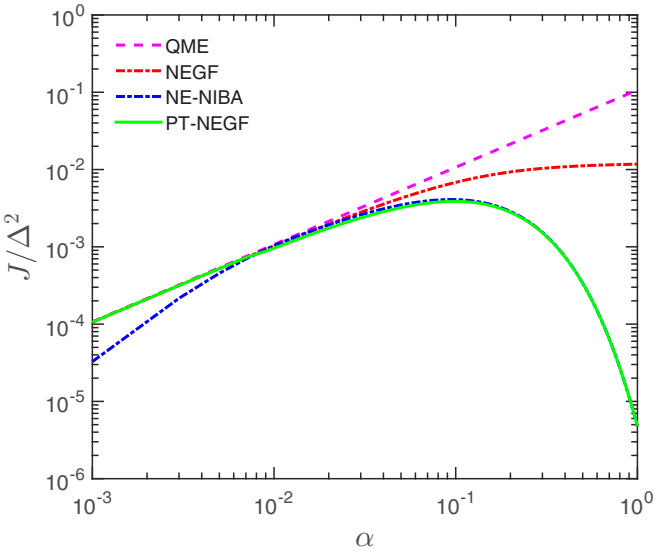


FIG. 1. Comparison between theoretical results on the energy current. The pink dashed line denotes the result of the QME [Eq. (42)], the blue dash-dotted line denotes the result of the NE-NIBA [Eq. (43)], the red dash-dotted line denotes the result of a previous NEGF method [Eq. (41)], and the green solid line denotes the result of the present PT-NEGF method [Eq. (7) together with Eq. (36)]. Parameters are $T_L/\Delta = 1.4$, $T_R/\Delta = 1.2$, $\alpha_L = \alpha_R = \alpha$, $\omega_c/\Delta = 30$ such that the nonadiabatic limit is fulfilled.

the NIBA framework for unbiased spin systems as has been noted in equilibrium cases [37]. These features indicate that our theory indeed provides a comprehensive and unified interpretation for energy transfer over a wide range of the coupling strength, a considerable improvement over existing theories.

Although our heat current formula can be applied to arbitrary temperature differences, it is still of primary interest to look at the behaviors in the linear response regime where some exact results can be adopted for comparison. We first consider the influence of the energy spacing $\Delta$ of the spin system on the energy conductance. According to the definition, $\kappa$ is totally determined by $\tilde{\chi}_z''(\omega)|_{T_L=T_R=T}$; in order to evaluate it, we only need Laplace transforms of bath correlations [Eq. (23)] at equilibrium states since all Fourier transforms can be expressed as Laplace transforms. In the scaling limit of $\omega_c/T \gg 1$, we have [38,45]

$$\Phi_{+-}(\lambda) = \frac{\Delta^2}{4\omega_c} e^{-i\pi\alpha} \frac{\Gamma(1-2\alpha)\Gamma(\alpha+|,\lambda/2\pi T)}{\Gamma(1-\alpha+\lambda/2\pi T)} \left(\frac{2\pi T}{\omega_c}\right)^{2\alpha-1} = \tilde{\Phi}^*_{+-}(\lambda). \tag{44}$$

Results for $\kappa$ depicted in Fig. 2 show a turnover behavior as a function of the energy spacing $\Delta$, which is in accordance with the findings of the multilayer multiconfiguration time-dependent Hartree theory [30] as well as the influence functional path integral method [15]. This dependence is due to the resonant character of energy transfer. The energy transfer is most efficient if the energy scale of the bridge subsystem is comparable to the temperature of the bath, as can be found that the value of $\Delta$ corresponding to the turnover point for $T/\omega_c = 0.02$ is almost double that for $T/\omega_c = 0.01$.

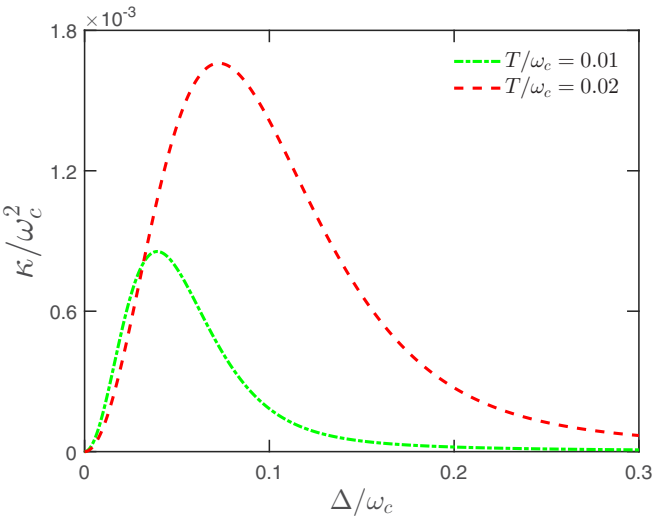


FIG. 2. Dependence of the energy conductance on the energy spacing $\Delta$ of the spin system for different temperatures. We choose $\alpha = 0.1$, $T_L = T_R = T$.

We next consider the temperature dependence of the energy conductance as shown in Fig. 3. In the weak coupling regime [Fig. 3(a)], we found our results match very well with those of the fully harmonic thermal junction, which has a simple harmonic oscillator (SHO) as the intermediate system [42] in the very low temperature regime. This is expected since at the low temperature limit the SHO is generally confined to its two lowest energy levels, thus it behaves like a two-level system [21,47]. When increasing the temperature, our formula reduces to the QME as demonstrated before. In the strong coupling regime, we consider the so-called Toulouse point with $\alpha = 0.5$ as an example, since the exact expression for equilibrium $\tilde{\chi}_z''(\omega)$ exists, which reads [38]

$$\tilde{\chi}_z''(\omega) = \mathrm{Im}\left[\frac{2}{\pi\omega}\frac{\gamma}{\omega+i\gamma}\Psi(\omega)\right], \tag{45}$$

where $\gamma = \pi\Delta^2/(2\omega_c)$, $\Psi(\omega) = 2\psi(x) - 2\psi(x - i\beta\omega/2\pi)$ with $\psi$ the digamma function and $x = \frac{1}{2}(1 + \frac{1}{2}\gamma\beta)$. As can be

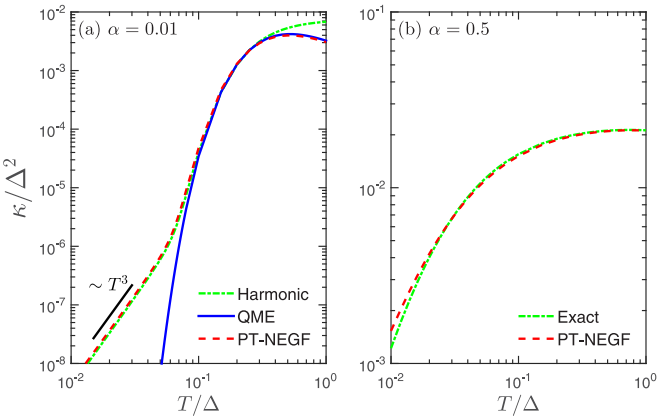


FIG. 3. Temperature dependence of the energy conductance with (a) $\alpha = 0.01$ and (b) $\alpha = 0.5$ (Toulouse point). We choose $\omega_c/\Delta = 20$, $T_L = T_R = T$.

seen from Fig. 3(b), our perturbation theory can approximate the exact results in a wide range of temperature; deviations only appear in the extreme low temperature regime due to the shortcomings of the PT [35]. Nevertheless, from the figure we clearly demonstrate that our theory is valid in a wide range of temperature in both the weak and strong system-bath coupling regime.

### B. Biased system

In this subsection we turn to the biased spin system. Although its dissipative dynamics has been extensively studied [9,38,48–52], a thorough understanding of its energy transfer characteristic is limited to the time-dependent case [33]. So far, no numerical methods could address effects of a finite bias at steady states. On the theoretical side, only the QME and NE-NIBA can involve bias in their formula [15]. However, their validity regimes are limited. It is then of necessity and interest to explore the energy transfer behaviors of the NESB model with a finite bias using our approach.

For biased systems, the QME framework predicted that [15]

$$J_Q = \frac{\Delta^2}{\omega_0}\frac{I_L(\omega_0)I_R(\omega_0)[n_L(\omega_0)-n_R(\omega_0)]}{I_L(\omega_0)[1+2n_L(\omega_0)]+I_R(\omega_0)[1+2n_R(\omega_0)]} \quad (46)$$

with $\omega_0 \equiv \sqrt{\Delta^2+\varepsilon^2}$ the energy spacing of the spin. While the energy current of the NE-NIBA reads [26,27]

$$J_N = \frac{1}{2\pi}\int_{-\infty}^{+\infty} \omega d\omega [P_1 C_R(\omega)C_L(\varepsilon-\omega) - P_0 C_R(-\omega)C_L(\omega-\varepsilon)], \quad (47)$$

where $C_v(\omega)$ is the Fourier transform of Eq. (37), $P_{0,1}$ denote the steady-state population of the spin states which are determined by the Fourier transform of bath correlation functions [Eq. (38)]:

$$P_0 = \frac{\Phi_{+-}(\varepsilon)}{\Phi_{+-}(\varepsilon)+\tilde{\Phi}_{+-}(\varepsilon)},$$
$$P_1 = \frac{\tilde{\Phi}_{+-}(\varepsilon)}{\Phi_{+-}(\varepsilon)+\tilde{\Phi}_{+-}(\varepsilon)}. \quad (48)$$

Results for the energy current are shown in Fig. 4. From the figure, we found that the PT-NEGF scheme still agrees with the QME in the weak coupling regime. Increasing the coupling strength, the QME predicts an almost linearly increasing $\kappa$ which is qualitatively incorrect, whereas our results and the NE-NIBA show turnover behaviors, although the turnover point differs. In the strong coupling regime, the PT-NEGF approaches the NE-NIBA. Thus the PT-NEGF can describe the energy transfer in biased systems.

In the intermediate coupling regime, we further notice an interesting phenomenon that the energy conductance is a nonmonotonic function of the bias as can be seen from Fig. 5. Such a dependence is directly in contrast to the NE-NIBA's prediction $\kappa \propto \varepsilon/\sinh(\varepsilon/T)$ for $\alpha = 0.1$–$0.5$ [15]. Note that for the spin system with fixed $\Delta$, the energy spacing $\omega_0$ increases as bias increases, so the nonmonotonic dependence of $\kappa$ on $\varepsilon$ at moderate couplings is similar to the results presented in Fig. 2. Therefore we can attribute this nonmonotonic behavior to the energy resonance between the bath and the spin. The NE-NIBA fails to capture this behavior,

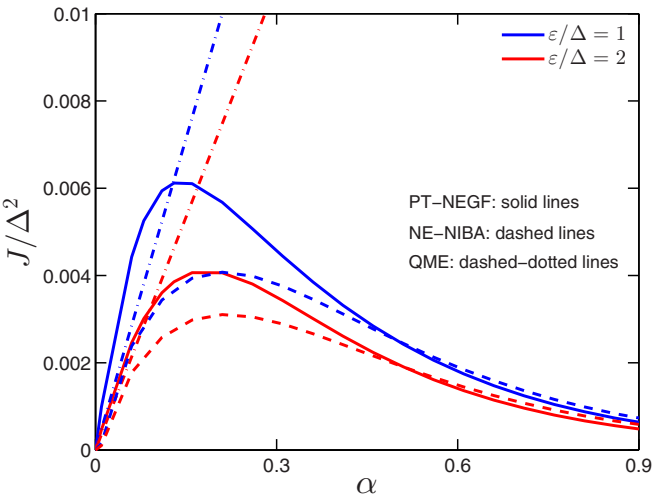


FIG. 4. Behaviors of the energy current with varying bias (blue denotes $\varepsilon = 1$, red denotes $\varepsilon = 2$) as a function of coupling strength. We compare PT-NEGF results (solid lines) to QME [Eq. (46)] (dash-dotted line) and NE-NIBA [Eq. (47)] (dashed lines). The inset shows the dependence of heat conductance on the bias for different coupling strengths predicted by the PT-NEGF. We choose $T_L/\Delta = 1.4$, $T_R/\Delta = 1.2$, $\alpha_L = \alpha_R = \alpha$, $\omega_c/\Delta = 30$ such that the nonadiabatic limit is fulfilled.

thus it is invalid in the intermediate coupling regime in the presence of bias; only in the strong coupling regime can we observe predicted monotonic behaviors of $\kappa$ as shown in the figure.

In accordance with such a nonmonotonic dependence, we would expect that if $\omega_0$ is always smaller (larger) than the bath temperature $T$, $\kappa$ should be a monotonically increasing (decreasing) function of $\varepsilon$ at moderate couplings. To verify this, we choose appropriate temperatures and vary bias. Results

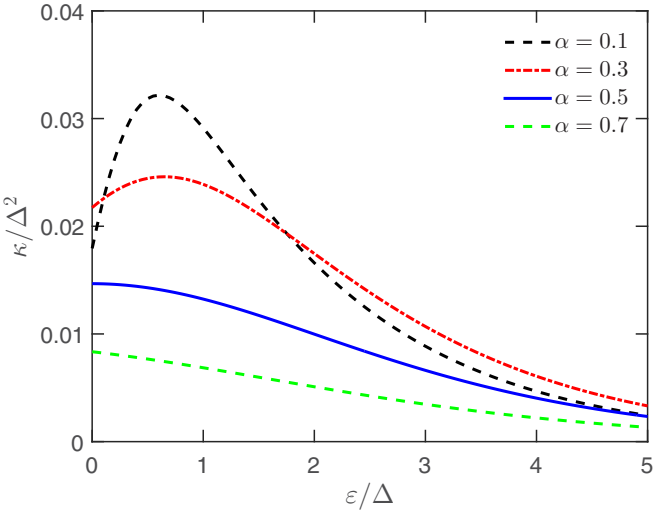


FIG. 5. The dependence of energy conductance on the bias for different coupling strengths predicted by the PT-NEGF. We choose $T_L/\Delta = T_R/\Delta = 1.2$, $\omega_c/\Delta = 30$ such that the nonadiabatic limit is fulfilled.

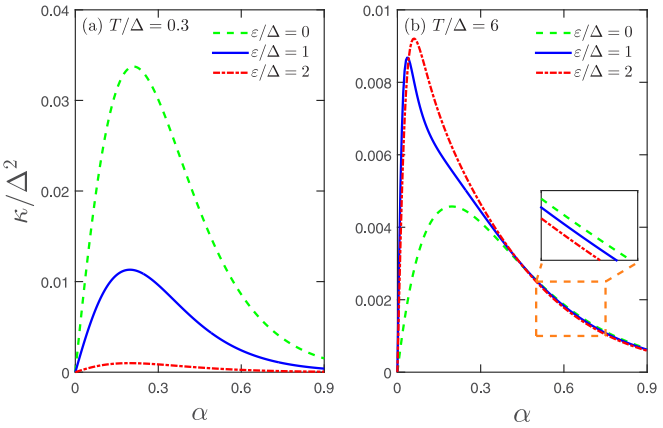


FIG. 6. Behaviors of the heat conductance with varying bias as a function of coupling strength for (a) $T/\Delta = 0.3$ and (b) $T/\Delta = 6$. The inset shows details of $\kappa$ in the strong coupling regime. We choose $T_L = T_R = T$ and $\omega_c/T = 30$ such that the nonadiabatic limit is fulfilled.

are shown in Fig. 6. As can be seen from the figure, the dependence of $\kappa$ on $\varepsilon$ in the intermediate coupling regime indeed meets our expectation. We also observe that $\kappa$ is always a monotonically decreasing function of $\varepsilon$ in the strong coupling regime, regardless of values of temperature, since this regime is dominated by the strong dissipation from the bath. Noting that values of the bias can be adjusted by changing the applied magnetic field, the interplay of the temperature, the bias, and the coupling strength may offer a nontrivial quantum control knob over heat transfer at the nanoscale. Our future work will address this aspect in more detail.

## V. SUMMARY

We formulate a PT-NEGF to investigate energy transfer in nonequilibrium spin-boson models. In contrast to previous NEGF methods treating the system-bath interaction as a perturbation, our PT-NEGF method treats the system-bath coupling nonperturbatively. Furthermore, in order to evaluate terms in the expansion series, we adopt the Majorana-fermion representation such that standard Feynman diagram techniques as well as the Dyson equation can be applied to spin systems. By doing so, our theoretical scheme goes beyond existing methods; it can tackle the strong coupling regime and include effects of a finite bias.

To demonstrate the utility of the approach, we first consider unbiased spin systems. Our analysis shows that the PT-NEGF method can give a comprehensive and unified interpretation for energy transfer over wide ranges of the coupling strength as well as temperature, a considerable improvement over existing theories. The predicted behavior of energy conductance as a function of the energy spacing of the spin is in accord with exact numerical simulations. When a finite bias plays a role, we found the energy conductance endows a nonmonotonic bias dependence at moderate coupling strengths, which is not reported in the present literature. We attribute this phenomenon to the resonant character of energy transfer in such systems. To verify our interpretation, we further consider different temperature regimes and obtain self-consistent results. These features make our theory stand out from previous considerations.

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