Inelastic cotunneling-induced decoherence and relaxation, charge, and spin currents in an interacting quantum dot under a magnetic field

Bing Dong
Department of Physics, Shanghai Jiaotong University, 1954 Huashan Road, Shanghai 200030, China and Department of Physics and Engineering Physics, Stevens Institute of Technology, Hoboken, New Jersey 07030, USA

Norman J. M. Horing
Department of Physics and Engineering Physics, Stevens Institute of Technology, Hoboken, New Jersey 07030, USA

H. L. Cui
Department of Physics and Engineering Physics, Stevens Institute of Technology, Hoboken, New Jersey 07030, USA and School of Optoelectronics Information Science and Technology, Yantai University, Yantai, Shandong, China
(Received 23 May 2005; revised manuscript received 27 July 2005; published 20 October 2005)

We present a theoretical analysis of several aspects of nonequilibrium cotunneling through a strong Coulomb-blockaded quantum dot (QD) subject to a finite magnetic field in the weak coupling limit. We carry this out by developing a generic quantum Heisenberg-Langevin equation approach leading to a set of Bloch dynamical equations which describe the nonequilibrium cotunneling in a convenient and compact way. These equations describe the time evolution of the spin variables of the QD explicitly in terms of the response and correlation functions of the free reservoir variables. This scheme not only provides analytical expressions for the relaxation and decoherence of the localized spin induced by cotunneling, but it also facilitates evaluations of the nonequilibrium magnetization, the charge current, and the spin current at arbitrary bias-voltage, magnetic field, and temperature. We find that all cotunneling events produce decoherence, but relaxation stems only from *inelastic* spin-flip cotunneling processes. Moreover, our specific calculations show that cotunneling processes involving electron transfer (both spin-flip and non-spin-flip) contribute to charge current, while spin-flip cotunneling processes are required to produce a net spin current in the asymmetric coupling case. We also point out that under the influence of a nonzero magnetic field, spin-flip cotunneling is an energy-consuming process requiring a sufficiently strong external bias-voltage for activation, explaining the behavior of differential conductance at low temperature: in particular, the splitting of the zero-bias anomaly in the charge current and a broad zero-magnitude "window" of differential conductance for the spin current near zero-bias-voltage.

DOI: 10.1103/PhysRevB.72.165326 PACS number(s): 73.63.Kv, 72.10.Fk, 72.15.Qm, 03.65.Yz

I. INTRODUCTION

Recent advances in probing and manipulating electronic spin in semiconductor quantum dots and other nanostructures hold the promise of new applications relating to quantum computation and quantum information processing. A single electronic spin not only can be used as an elementary quantum memory unit, i.e., the qubit, due to its relatively long relaxation time in semiconductors, but it is also expected to be useful as an element of calculation in the context of quantum computing algorithms or quantum information transport processing, which depend essentially on its temporal persistence of quantum interference.[1]

This expectation provides strong motivation to develop a full understanding of the coherent evolution dynamics of a single spin in semiconductors. Actually, much effort has been made on this matter from both theoretical and experimental points of view. In particular, for a single quantum dot (QD), it has been predicted that measurements of the tunneling current between two leads via this QD may be an appropriate experimental tool to extract information about the orientation and dynamics of a single spin localized inside the QD.[2] Indeed, recent scanning tunneling experiments have observed modulation of the tunneling current through a single molecule with a spin subject to a constant magnetic field at the Larmor frequency, which is the characteristic dynamical (precession) frequency of a single spin under influence of magnetic field, with a corresponding peak in its noise power spectrum.[3,4] This feature has been examined theoretically on the basis of two different weak tunneling models in the strong Coulomb blockade regime: (a) sequential tunneling using a simple quantum rate equation approach,[5] and (b) the two-channel Kondo Hamiltonian using the nonequilibrium Green's function formalism jointly with the Majorana-fermion representation.[6]

However, from a fundamental quantum mechanical point of view, any quantum measurement will inevitably introduce some *disturbances* into the measured system and consequently induce decoherence in the system variable conjugate to the one being measured. Therefore, the information concerning spin dynamics extracted from a tunneling measurement is expected to involve a reaction signature of the tunneling upon the coherent evolution of the single spin. It is therefore crucial to theoretically account for the tunneling-measurement-induced spin relaxation and decoherence behaviors as functions of temperature and bias-voltage applied between the two leads; so as to provide a better understanding of the information obtained from measurement, and to give useful insight on how to raise measurement efficiency.

The earlier papers cited above have concentrated on interpretation of the peak in the current noise spectrum, but a systematic investigation of the nonequilibrium relaxation and decoherence effects, as far as we know, is still lacking. It is the main purpose of this paper to perform these investigations. In treating the open quantum system at hand, we will employ the quantum Heisenberg-Langevin equation approach, to establish a set of quantum Bloch equations (i.e., equations of motion for the reduced density matrix) for a two-level system (a single spin in the QD) tunnel-coupled to two normal leads in a fully microscopic way, and then proceed to study the dynamics of a single spin qubit in an ambient magnetic field under nonequilibrium transport conditions.

As mentioned above, two different tunneling mechanisms have been utilized to describe the quantum measurement process. If the chemical potentials of the two leads are nearly matched with the energy level of the sandwiched spin, the resonance condition is satisfied and the lowest-order tunneling, i.e., sequential tunneling, is observed in the transport process. However, it is quite likely that the chemical potentials are probably further from the resonant point in actual experiments. In this case, the lowest-order tunneling of an electron into the QD is largely suppressed, but at very low temperature, a higher-order tunneling mechanism known as *inelastic cotunneling* dominates the transport in the strong Coulomb blockade regime; in this mechanism an electron tunnels from the left lead to a virtual state in the dot, and then another electron tunnels from the dot to the right lead without changing the charge inside the QD. This is the tunneling mechanism that gives rise to the Kondo effect in QD tunneling. In fact, an exact mapping has been established between such cotunneling and the anisotropic Kondo problem by analyzing and comparing their respective perturbation series for tunneling amplitudes.[7] Accordingly, we will adopt the Kondo Hamiltonian in this paper to describe the inelastic cotunneling process and study its dissipation involved in coherent tunneling via the QD: we do so by developing a generic Langevin equation approach in second-order perturbation theory with respect to the *s-d* exchange coupling constant, J, in the weak tunneling limit.

Moreover, we will evaluate the nonequilibrium spin magnetization of a QD subject to a magnetic field in steady state and examine the behavior of charge flow (cotunneling current) through the QD within the same framework. Actually, an interesting calculation of the spin magnetization of a Kondo QD has already been carried out recently by means of the Majorana-fermion Green's function technique.[8] The striking result obtained in that paper is that their magnetization result differs from the thermal equilibrium formula even at zero order in the spin-leads exchange coupling, J. More impressively, theoretical analysis shows that if proper account of this nonequilibrium magnetization is taken in a calculation of the current, the resulting differential conductance will demonstrate double peaks at bias voltages $eV=\pm g\mu_B B$ (the Zeeman energy), a signature of Kondo effect with a constant magnetic field B, even in the second-order perturbation calculations.[9] Of course, the log-signature peculiarity of the Kondo effect occurs essentially in the next orders of perturbation theory. Here, in the present paper, we ignore such higher-order terms; thus we confine our study to the ordinary cotunneling processes. A detailed analysis of the third-order perturbation contribution to the current has been established in Ref. 9, computing an explicit logarithmic enhancement in current.

Recent theoretical studies have shown that nonequilibrium Kondo physics is fundamentally governed by weak-tunneling perturbation theory when the bias voltage is much larger than the Kondo temperature, T_K; this can be ascribed to current induced decoherence of the resonant spin-flip term in cotunneling processes, which can eliminate the generic logarithmic divergence in conventional Kondo physics.[10–12] This is the reason that the third-order contribution in Ref. 9 provides a quantitatively relatively small modification to the second-order term in the nonequilibrium current formula in the weak tunneling limit (albeit qualitatively important). It is also shown in Ref. 9 that the second-order calculation (cotunneling) of the differential conductance exhibits cusps at bias voltages $eV=\pm g\mu_B B$, a remnant of the Kondo effect. Therefore, we call this cotunneling behavior "Kondo-type" cotunneling. In this paper, we will systematically study this kind of nonequilibrium cotunneling through a single spin in a finite magnetic field using second-order perturbation theory and will specifically analyze the cotunneling processes responsible for its special transport characteristics.

In addition, we will also examine the behavior of the spin current and show that *inelastic* spin-flip cotunneling can produce a nonzero spin current for asymmetric coupling systems subject to a finite magnetic field and an activation bias-voltage. Unlike charge current, we find that the sign of spin current is independent of the direction of the applied bias-voltage, but it does depend on the asymmetry of the coupling constants to the left and right leads and the direction of external magnetic field.

The remaining parts of the paper are organized as follows. In Sec. II, we present the physical model used in this paper: a single spin weakly tunnel-coupled to two normal leads. To focus on tunneling induced decoherence here, it is assumed that the single spin is free of any other dissipative heat bath except for the tunneling reservoirs. In Sec. III, we will then present the derivation of the quantum Langevin equations of motion for the single spin. In Sec. IV, qualitative discussion and concrete calculations will be given for the resulting decoherence and relaxation rates as functions of magnetic field, bias-voltage, and temperature. Section V focuses first on the derivation of closed-form expressions for charge current and spin current within the framework of the quantum Langevin equation approach developed here, and then addresses all possible cotunneling processes that occur in this system, and their respective contributions to the currents. A numerical evaluation of differential conductance for the charge current and the spin current is provided in the last part of this section. Finally, our conclusions are summarized in Sec. VI.

II. MODEL HAMILTONIAN

We employ the two-lead Kondo Hamiltonian discussed above to model inelastic cotunneling through a single spin (or QD) in a magnetic field, B, in the weak-coupling regime:

$$H = H_0 + H_{\rm I}, \quad H_{\rm I} = H_{\rm refl} + H_{\rm trans},$$

$$H_0 = \sum_{\eta\mathbf{k}\sigma} (\varepsilon_{\mathbf{k}} - \mu_\eta) c^\dagger_{\eta\mathbf{k}\sigma} c_{\eta\mathbf{k}\sigma} - g\mu_B B S^z,$$

$$H_{\rm refl} = \sum_{\eta,\mathbf{k},\mathbf{k}'} J_{\eta\eta}[(c^\dagger_{\eta\mathbf{k}\uparrow} c_{\eta\mathbf{k}'\uparrow} - c^\dagger_{\eta\mathbf{k}\downarrow} c_{\eta\mathbf{k}'\downarrow})S^z + c^\dagger_{\eta\mathbf{k}\uparrow} c_{\eta\mathbf{k}'\downarrow} S^- + c^\dagger_{\eta\mathbf{k}\downarrow} c_{\eta\mathbf{k}'\uparrow} S^+],$$

$$H_{\rm trans} = J_{RL} \sum_{\mathbf{k},\mathbf{k}'} [(c^\dagger_{R\mathbf{k}\uparrow} c_{L\mathbf{k}'\uparrow} - c^\dagger_{R\mathbf{k}\downarrow} c_{L\mathbf{k}'\downarrow})S^z + c^\dagger_{R\mathbf{k}\uparrow} c_{L\mathbf{k}'\downarrow} S^- + c^\dagger_{R\mathbf{k}\downarrow} c_{L\mathbf{k}'\uparrow} S^+] + (R \leftrightarrow L), \tag{1}$$

where $c^\dagger_{\eta\mathbf{k}\sigma}$ ($c_{\eta\mathbf{k}\sigma}$) creates (annihilates) an electron in lead η (=L,R) with momentum $\mathbf{k}$, spin σ and bare energy $\varepsilon_{\mathbf{k}}$. J_{LL}, J_{RR}, and $J_{LR}=J_{RL}=\sqrt{J_{LL}J_{RR}}$ are Kondo exchange coupling constants between the electrons and the localized spin-$\frac{1}{2}$, $\mathbf{S}=(S^x,S^y,S^z)$, $S^\pm = S^x \pm iS^y$. H_0 stands for the free Hamiltonian containing (1) two noninteracting normal leads, individually in local equilibrium with temperature T (not to be confused with decay times to be introduced below), respective chemical potentials μ_η, and Fermi distribution functions defined as $f_\eta(\epsilon)=[1+e^{(\epsilon-\mu_\eta)/k_BT}]^{-1}$; and (2) Zeeman energy of the localized spin subject to magnetic field B (g and μ_B are the Landé factor and the Bohr magneton, respectively). It should be noted that we ignore the Zeeman effect in the lead electrons. The interaction part of the total Hamiltonian, $H_{\rm I}$, also includes two terms: $H_{\rm refl}$ describes the reflection processes, in which an electron from a given lead is scattered back into the *same* lead in both spin-conserving and spin-flip configurations; while $H_{\rm trans}$ describes the transmission events, where an electron from one lead cotunnels into the *other* lead, also in both configurations. Except for the tunnel coupling, we ignore all other "environmental" decay interactions of the single spin.

Here, we assume the leads to have a flat density of states, ρ_η, in the wide-band limit. We take the chemical potentials, μ_η, to vanish in equilibrium and use this choice as the reference of energy throughout the paper. In the nonequilibrium case, we assume the bias-voltage is applied symmetrically, $\mu_L=-\mu_R=eV/2$. Throughout, we will use units with $\hbar=k_B=e=1$.

The conceptual structure of our model is predicated on the idea that the full system can be separated into two subsystems: one of which is the measured subsystem, the single spin, and the other consists of the two leads jointly comprising a "heat bath" or "reservoir." The interaction between the two subsystems, $H_{\rm I}$, must be weak in order that the separation of the two subsystems is physically meaningful. Accordingly, $H_{\rm I}$ generates dissipation for the dynamics of the "open" measured quantum subsystem, which is the principal focus for study in this paper. For notational brevity, we rewrite this term as a sum of three products of two variables:

$$H_{\rm I} = Q^z F_{Q^z} + Q^+ F_{Q^+} + Q^- F_{Q^-}, \tag{2a}$$

with

$$Q^z = \sum_{\eta,\eta'} Q^z_{\eta\eta'} = \sum_{\eta,\eta',\mathbf{k},\mathbf{k}'} J_{\eta\eta'}(c^\dagger_{\eta\mathbf{k}\uparrow} c_{\eta'\mathbf{k}'\uparrow} - c^\dagger_{\eta\mathbf{k}\downarrow} c_{\eta'\mathbf{k}'\downarrow}), \tag{2b}$$

$$Q^+ = \sum_{\eta,\eta'} Q^+_{\eta\eta'} = \sum_{\eta,\eta',\mathbf{k},\mathbf{k}'} J_{\eta\eta'} c^\dagger_{\eta\mathbf{k}\uparrow} c_{\eta'\mathbf{k}'\downarrow}, \tag{2c}$$

$$Q^- = \sum_{\eta,\eta'} Q^-_{\eta\eta'} = \sum_{\eta,\eta',\mathbf{k},\mathbf{k}'} J_{\eta\eta'} c^\dagger_{\eta\mathbf{k}\downarrow} c_{\eta'\mathbf{k}'\uparrow}, \tag{2d}$$

as functions of reservoir variables, and the corresponding generalized forces, F_Q, depend on the variables of the measured subsystem as

$$F_{Q^z} = S^z, \quad F_{Q^+} = S^-, \quad F_{Q^-} = S^+. \tag{2e}$$

Here, the terms $Q^\pm F_{Q^\pm}$ describe spin-flip tunneling processes, in which the conduction electron spin changes its orientation in the process of tunneling, and the localized spin is also flipped. On the other hand, the term $Q^z F_{Q^z}$ is responsible for non-spin-flip (spin-conserving) tunneling, in which no spin exchange takes place. All of these tunneling processes are schematically elaborated in Fig. 3.

III. QUANTUM LANGEVIN EQUATIONS

In this section, we derive a generic quantum Langevin equation approach and establish a set of quantum Bloch equations to describe the dynamics of a single spin modeled by Eq. (1). It is well known that the underlying quantum Langevin equation approach has been extensively developed and successfully employed in the contexts of quantum electrodynamics and quantum optics.[13–15] Albeit that the great advantage of this scheme is that it allows us to naturally incorporate the effects of quantum noise introduced by the "environment" on the studied system variables,[16] we will take no account of such fluctuation issues here. Considering that such noise has a very short correlation time (determined by the reservoir correlation time, τ_r), it is reasonable to neglect it for the longer time scale ($>\tau_r$) of interest in the present paper.

The Heisenberg equations of motion for the spin Pauli operators $S^z, S^\pm$ and the lead operators are given by

$$i\dot{S}^z = [S^z, H]_- = :[S^z, H_{\rm I}]_-: = :(Q^- S^+ - Q^+ S^-):, \tag{3}$$

$$i\dot{S}^\pm = [S^\pm, H]_- = \pm\Delta S^\pm + :[S^\pm, H_{\rm I}]_-: = \pm\Delta S^\pm \pm :(2Q^\pm S^z - Q^z S^\pm):, \tag{4}$$

$$i\dot{c}_{\eta\mathbf{k}\uparrow} = [c_{\eta\mathbf{k}\uparrow}, H]_- = \varepsilon_{\eta\mathbf{k}} c_{\eta\mathbf{k}\uparrow} + :[c_{\eta\mathbf{k}\uparrow}, H_{\rm I}]_-: = \varepsilon_{\eta\mathbf{k}} c_{\eta\mathbf{k}\uparrow} + \sum_{\mathbf{k}'} [S^z(J_{\eta\eta} c_{\eta\mathbf{k}'\uparrow} + J_{\eta\bar{\eta}} c_{\bar{\eta}\mathbf{k}'\uparrow}) + S^-(J_{\eta\eta} c_{\eta\mathbf{k}'\downarrow} + J_{\eta\bar{\eta}} c_{\bar{\eta}\mathbf{k}'\downarrow})], \tag{5}$$

$$i\dot{c}_{\eta\mathbf{k}\downarrow}=[c_{\eta\mathbf{k}\downarrow},H]_-=\varepsilon_{\eta\mathbf{k}}c_{\eta\mathbf{k}\downarrow}+:[c_{\eta\mathbf{k}\downarrow},H_{\mathrm{I}}]_-:$$
$$=\varepsilon_{\eta\mathbf{k}}c_{\eta\mathbf{k}\downarrow}+\sum_{\mathbf{k}'}[-S^z(J_{\eta\eta}c_{\eta\mathbf{k}'\downarrow}+J_{\eta\bar{\eta}}c_{\bar{\eta}\mathbf{k}'\downarrow})$$
$$+S^-(J_{\eta\eta}c_{\eta\mathbf{k}'\uparrow}+J_{\eta\bar{\eta}}c_{\bar{\eta}\mathbf{k}'\uparrow})], \quad (6)$$

where we have $\varepsilon_{\eta\mathbf{k}}=\varepsilon_{\mathbf{k}}-\mu_\eta$, $\bar{\eta}=L(R)$ if $\eta=R(L)$, and $\Delta=g\mu_B B$. $[A,B]_\pm\equiv AB\pm BA$ are, respectively, the commutator and the anticommutator of operators A and B. The equations of motion for $c^\dagger_{\eta\mathbf{k}\sigma}$ are easily obtained by Hermitian conjugation of the equations for $c_{\eta\mathbf{k}\sigma}$. The colon-pair notation, $:(\cdots):$, in these equations denotes *normal ordering* of the operators, $\cdots$, inside the square brackets: all annihilation reservoir operators $c_{\eta\mathbf{k}\sigma}$ are placed to the right of all spin operators, $S^{z(\pm)}$, and the creation reservoir operators $c^\dagger_{\eta\mathbf{k}\sigma}$ are placed to the left of all spin operators, if the operators involved have equal-time arguments. For instance, the last two lines in Eqs. (5) and (6) are already normal-ordered. This normal ordering employed here is an operator counterpart of determining a cumulant in terms of Feynman diagrams with the elimination of disconnected diagrams involving products of lower order Green's functions. The latter disconnected diagram terms involve the effects of weak coupling (to the bath) which oscillate rapidly at the high frequencies of microscopic dynamics, with attendant destructive interference. While such terms do contribute small quantum corrections ("renormalization," "radiative corrections") to the microscopic dynamics on a short time scale, they are negligible in the context of the much longer time scale implicitly under consideration in our formulation of a quantum Heisenberg-Langevin equation. A full explanation of the normal ordering scheme in the equations of motion is provided in Refs. 13–17, to which we refer the reader.

Formally integrating these Heisenberg equations of motion from initial time 0 to t we obtain the exact solutions for these operators as

$$S^z(t)=S^z(0)-i\int_0^t dt':[S^z(t'),H_{\mathrm{I}}(t')]_-:, \quad (7a)$$

$$S^\pm(t)=e^{\mp i\Delta t}S^\pm(0)-i\int_0^t dt' e^{\mp i\Delta(t-t')}:[S^\pm(t'),H_{\mathrm{I}}(t')]_-:, \quad (7b)$$

$$c_{\eta\mathbf{k}\sigma}(t)=e^{-i\varepsilon_{\eta\mathbf{k}}t}c_{\eta\mathbf{k}\sigma}(0)-i\int_0^t dt' e^{-i\varepsilon_{\eta\mathbf{k}}(t-t')}$$
$$\times :[c_{\eta\mathbf{k}\sigma}(t'),H_{\mathrm{I}}(t')]_-:. \quad (7c)$$

In the absence of interaction, $H_{\mathrm{I}}\to 0$, these solutions become

$$S^z_o(t)=S^z_o(t'), \quad (8a)$$

$$S^\pm_o(t)=e^{\mp i\Delta(t-t')}S^\pm_o(t'), \quad (8b)$$

$$c^o_{\eta\mathbf{k}\sigma}(t)=e^{-i\varepsilon_{\eta\mathbf{k}}(t-t')}c^o_{\eta\mathbf{k}\sigma}(t'). \quad (8c)$$

A standard assumption in the derivation of a quantum Langevin equation is that the time scale of decay processes is much slower than that of free evolution, which is reasonable in the weak-tunneling approximation. In this context it is appropriate to substitute the time-dependent decoupled reservoir and spin operators of Eqs. (8a)–(8c) into the formal solutions of Eqs. (7a)–(7c). Obviously, the full solution for the reservoir operator comprises two contributions, one from free evolution and the other from reaction of the spin through the weak coupling, and we denote these with superscripts "o" and "i," respectively:

$$c_{\eta\mathbf{k}\sigma}(t)=c^o_{\eta\mathbf{k}\sigma}(t)+c^i_{\eta\mathbf{k}\sigma}(t), \quad (9a)$$

with

$$c^i_{\eta\mathbf{k}\sigma}(t)=-i\int_0^t dt':[c^o_{\eta\mathbf{k}\sigma}(t),H^o_{\mathrm{I}}(t')]_-:, \quad (9b)$$

where H^o_{I} is composed of the operators in H_{I} which are replaced by their decoupled counterparts (interaction picture). In fact, this is just the operator formulation of linear response theory. It should also be noted that Eq. (9a) implies that the two subsystems, the quantum dot and the reservoirs, are completely isolated before $t_0=0$, and the perturbative interaction, H_{I}, is adiabatically switched on from the initial time $t=t_0$. Using Eqs. (9a) and (9b), the reservoir variables, $Q^{z(\pm)}_{\eta\eta'}(t)$, become (Appendix A)

$$Q^z_{\eta\eta'}(t)=Q^{zo}_{\eta\eta'}(t)+Q^{zi}_{\eta\eta'}(t)$$
$$=Q^{zo}_{\eta\eta'}(t)-i\theta(\tau)\int_{-\infty}^t d\tau:[Q^{zo}_{\eta\eta'}(t),H^o_{\mathrm{I}}(t')]_-:, \quad (10a)$$

$$Q^\pm_{\eta\eta'}(t)=Q^{\pm o}_{\eta\eta'}(t)+Q^{\pm i}_{\eta\eta'}(t)$$
$$=Q^{\pm o}_{\eta\eta'}(t)-i\theta(\tau)\int_{-\infty}^t d\tau:[Q^{\pm o}_{\eta\eta'}(t),H^o_{\mathrm{I}}(t')]_-:, \quad (10b)$$

with $\tau=t-t'$ and $\theta(\tau)$ represents the Heaviside step-function. Similarly, the formal solutions for the spin operators are also divided into two parts:

$$S^z(t)=S^z_o(t)+S^z_i(t)=S^z_o(t)-i\theta(\tau)\int_{-\infty}^t d\tau:[S^z_o(t),H^o_{\mathrm{I}}(t')]_-:, \quad (11a)$$

$$S^\pm(t)=S^\pm_o(t)+S^\pm_i(t)=S^\pm_o(t)-i\theta(\tau)\int_{-\infty}^t d\tau:[S^\pm_o(t),H^o_{\mathrm{I}}(t')]_-:. \quad (11b)$$

Substituting these approximate solutions of Eqs. (10a), (10b), (11a), and (11b) into the equations of motion for S^z and $S^\pm$ [Eqs. (3) and (4)] and taking average evaluations with respect to the reservoir electron ensemble $\langle\cdots\rangle_e$ and over the localized spin degrees of freedom $\langle\cdots\rangle_s$, one can derive the desired quantum Bloch equations up to second order in the Kondo coupling constant J. After some algebraic manipulations (details are provided in Appendix A), the quantum dynamic equations take the compact form:

$$\begin{aligned}\langle\dot{S}^z\rangle = &-\frac{1}{2}\theta(\tau)\int_{-\infty}^{t} d\tau\langle[Q_o^-(t),Q_o^+(t')]_+\rangle_e\langle[S^+(t),F_{Q^+}(t')]_-\rangle_s\\ &-\frac{1}{2}\theta(\tau)\int_{-\infty}^{t} d\tau\langle[Q_o^-(t),Q_o^+(t')]_-\rangle_e\langle[S^+(t),F_{Q^+}(t')]_+\rangle_s\\ &+\frac{1}{2}\theta(\tau)\int_{-\infty}^{t} d\tau\langle[Q_o^+(t),Q_o^-(t')]_+\rangle_e\langle[S^-(t),F_{Q^-}(t')]_-\rangle_s\\ &+\frac{1}{2}\theta(\tau)\int_{-\infty}^{t} dt'\langle[Q_o^+(t),Q_o^-(t')]_-\rangle_e\langle[S^-(t),F_{Q^-}(t')]_+\rangle_s,\end{aligned} \tag{12}$$

$$\begin{aligned}\langle\dot{S}^\pm\rangle = &\mp i\Delta\langle S^\pm\rangle \mp \theta(\tau)\int_{-\infty}^{t} d\tau\langle[Q_o^\pm(t),Q_o^\mp(t')]_+\rangle_e\\ &\times\langle[S^z(t),F_{Q^\mp}(t')]_-\rangle_s \mp \theta(\tau)\int_{-\infty}^{t} d\tau\langle[Q_o^\pm(t),Q_o^\mp(t')]_-\rangle_e\\ &\times\langle[S^z(t),F_{Q^\mp}(t')]_+\rangle_s \pm \frac{1}{2}\theta(\tau)\int_{-\infty}^{t} d\tau\langle[Q_o^z(t),Q_o^z(t')]_+\rangle_e\\ &\times\langle[S^\pm(t),F_{Q^z}(t')]_-\rangle_s \pm \frac{1}{2}\theta(\tau)\int_{-\infty}^{t} d\tau\langle[Q_o^z(t),Q_o^z(t')]_-\rangle_e\\ &\times\langle[S^\pm(t),F_{Q^z}(t')]_+\rangle_s.\end{aligned} \tag{13}$$

In these equations, we drop the superscript "o" in the spin operators occurring inside integrations, since they involve the dynamical spin variables after taking expectation values. However, it must be borne in mind that their time evolutions are governed by Eqs. (8a) and (8b). Apart from free evolution, it is clear that the spin dynamics are modified by the spin-lead interaction in a way that is precisely relevant to the response function, $R^{ab}(t,t')$, and correlation function, $C^{ab}(t,t')$, $(a,b=z,+,-)$ of free reservoir variables, which are defined as

$$R^{ab}(t,t') = \tfrac{1}{2}\theta(\tau)\langle[Q_o^a(t),Q_o^b(t')]_-\rangle_e, \tag{14}$$

$$C^{ab}(t,t') = \tfrac{1}{2}\theta(\tau)\langle[Q_o^a(t),Q_o^b(t')]_+\rangle_e. \tag{15}$$

These forms of quantum Langevin-type dynamic equations, expressed explicitly in terms of the correlation and response functions of free reservoir variables, have also been proposed in Ref. 18 by employing the quantum Furutsu-Novikov theorem. The present derivation seems more direct and its meaning is more transparent.

Considering the reservoirs to be in separate (local) equilibrium states except for differing chemical potentials (reflecting the bias-voltage driving the current) and noting that these free fermion reservoir operators, $c^o_{\eta\mathbf{k}\sigma}$, $c^{o\dagger}_{\eta\mathbf{k}\sigma}$, obey Wick's theorem (without correlation between the leads), we can readily express the functions $R^{ab}(t,t')$ and $C^{ab}(t,t')$ in terms of reservoir distribution functions. The calculational details are provided in Appendix B. Here we cite some useful properties. First, these response and correlation functions are functions only of the time difference $\tau=t-t'$. Second, these functions are related as

$$R(\tau) = R^{+-}(\tau) = R^{-+}(\tau) = \tfrac{1}{2}R^{zz}(\tau), \tag{16}$$

$$C(\tau) = C^{+-}(\tau) = C^{-+}(\tau) = \tfrac{1}{2}C^{zz}(\tau). \tag{17}$$

Therefore, it is convenient to introduce single Fourier time transforms for the two bath functions into frequency space:

$$R(\omega) = \int_{-\infty}^{\infty} d\tau e^{i\omega\tau}R(\tau), \tag{18}$$

$$C(\omega) = \int_{-\infty}^{\infty} d\tau e^{i\omega\tau}C(\tau). \tag{19}$$

Third, the spectral function $C(\omega)$ is an even function of ω, while the imaginary part of the frequency-dependent retarded susceptibility $R(\omega)$ is an odd function. In equilibrium, they are exactly related by the fluctuation-dissipation theorem.

Employing the definitions of response and correlation functions and free evolution relation $S^\pm(t')=e^{\pm i\Delta\tau}S^\pm(t)$, Eq. (12) yields

$$\begin{aligned}\dot{S}^z = &-2S^z\int_{-\infty}^{t} d\tau e^{-i\Delta\tau}C(\tau) - \int_{-\infty}^{t} d\tau e^{-i\Delta\tau}R(\tau)\\ &-2S^z\int_{-\infty}^{t} d\tau e^{i\Delta\tau}C(\tau) + \int_{-\infty}^{t} d\tau e^{i\Delta\tau}R(\tau).\end{aligned} \tag{20}$$

(Hereafter, we suppress the brackets around the spin variables since they are all c numbers.) In a transport measurement experiment, a single spin decays to its external bias-voltage-driven steady state in a characteristic time, τ_c, of the system. If we assume that the single spin changes significantly only over a time scale $t \gg \tau_c$, an appropriate Markov approximation may be generated by making the replacement

$$\int_{-\infty}^{t} d\tau \Rightarrow \int_{-\infty}^{\infty} d\tau. \tag{21}$$

From a physical point of view, this presumption is consistent with the normal ordering scheme performed in the operator equations of motion, in regard to elimination of the rapid oscillations in microscopic dynamics. In this case, the equation of motion for S^z can be further simplified as

$$\begin{aligned}\dot{S}^z &= -2[C(\Delta)+C(-\Delta)]S^z + R(\Delta) - R(-\Delta)\\ &= -4C(\Delta)S^z + 2R(\Delta).\end{aligned} \tag{22}$$

Analogously, the quantum Langevin equation for $S^\pm$ becomes

$$\begin{aligned}\dot{S}^\pm &= \mp i\Delta\ S^\pm - 2S^\pm\int_{-\infty}^{t} d\tau e^{\pm i\Delta\tau}C(\tau) - 2S^\pm\int_{-\infty}^{t} d\tau C(\tau)\\ &= \mp i\Delta\ S^\pm - 2[C(\Delta)+C(0)]S^\pm.\end{aligned} \tag{23}$$

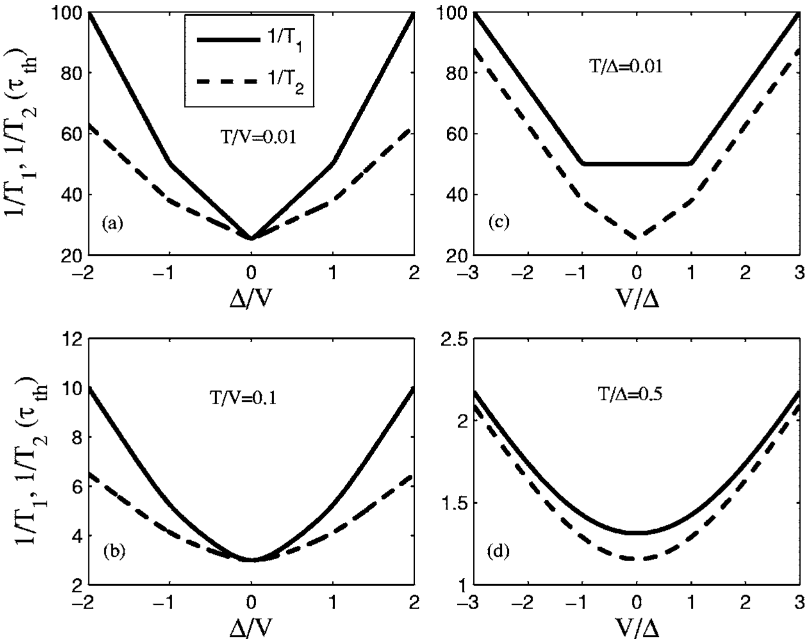

FIG. 1. The cotunneling-induced spin relaxation rate (T_1^{-1}) and decoherence rate (T_2^{-1}) as functions of magnetic field Δ [(a), (b)] and of bias-voltage V [(c), (d)] for given temperatures indicated in these figures. The parameters we use in calculation are $J_{LL}\rho_L = J_{RR}\rho_R = J_{LR}\sqrt{\rho_L\rho_R} = 0.02$. Here, we use the decay rate of purely thermal fluctuations, $\tau_{\rm th}$, Eq. (29), as the unit of the two rates.

IV. NONEQUILIBRIUM MAGNETIZATION, DECOHERENCE, AND RELAXATION

In this section, on the basis of the derived Bloch equations, Eqs. (22) and (23), we will carry out analytical evaluations of the relaxation and decoherence rates, as well as the magnetization of the single spin under transport conditions, as functions of temperature, bias-voltage, and external magnetic field.

It is well known that there are two decay mechanisms leading to standard Bloch equations which define two distinct relaxation time scales: (1) The longitudinal relaxation time, T_1, is responsible for the spin magnetic moment relaxation, while (2) the transverse relaxation time, T_2, is responsible for decoherence of the quantum superposition state composed of the two spin states $\sigma = \uparrow$ and $\downarrow$. These time scales are defined by the time evolutions of $S^z(t)$ and $S^{\pm}$, respectively:

$$\frac{1}{T_1} = 4C(\Delta) = 2\pi\left(J_{LL}^2\rho_L^2 + J_{RR}^2\rho_R^2\right)T\varphi\left(\frac{\Delta}{T}\right) + 2\pi J_{LR}^2\rho_L\rho_R T \times\left[\varphi\left(\frac{\Delta+V}{T}\right) + \varphi\left(\frac{\Delta-V}{T}\right)\right], \tag{24}$$

$$\frac{1}{T_2} = 2[C(\Delta) + C(0)] = \frac{1}{2T_1} + 2C(0) = \frac{1}{2T_1} + 2\pi(J_{LL}^2\rho_L^2 + J_{RR}^2\rho_R^2)T + 2\pi J_{LR}^2\rho_L\rho_R T\varphi\left(\frac{V}{T}\right). \tag{25}$$

In deriving these results, we employed Eqs. (B19) and (B20). It is noteworthy that the transverse spin relaxation rate, $1/T_2$, includes two contributions: the relaxation-induced dephasing, $1/2T_1$, and also pure decoherence, $2C(0)$.

From Eqs. (12) and (13), we can easily deduce that the longitudinal relaxation time (T_1) stems completely from spin-flip cotunneling events, which is conceptually consistent with the physical definition of spin relaxation and implies its dependence on magnetic field. Furthermore, spin-flip processes also contribute to decoherence with the partial rate, $1/2T_1$. In contrast, non-spin-flip processes do not induce spin relaxation but they do contribute to pure decoherence with the partial rate $2C(0)$, which is independent of magnetic field. This difference in the magnetic field dependences of the two rates may be understood in the following terms: the non-spin-flip process entails charge transport through the QD via a virtual state but the QD eventually returns back to its original spin state without changing energy (which is why this process is referred to as *elastic* cotunneling in the literature); whereas energy exchange does take place between the QD and leads in the spin-flip process in a finite magnetic field, in which the spin of the QD is finally flipped and thus the QD is *inelastically* excited or decays accompanied by excess energy, the Zeeman energy, Δ. Of course, in the absence of an external magnetic field, spin-flip cotunneling also becomes *elastic*. In this case, the two relaxation times are equal (i.e., it is inelastic cotunneling that makes them differ),

$$\frac{1}{T_1^0} = \frac{1}{T_2^0} = 4C(0) = 4\pi(J_{LL}^2\rho_L^2 + J_{RR}^2\rho_R^2)T + 4\pi J_{LR}^2\rho_L\rho_R T\varphi\left(\frac{V}{T}\right). \tag{26}$$

On the other hand, in the limit of zero bias-voltage (equilibrium condition), the relaxation rates become

$$\frac{1}{T_1^{\rm eq}} = 4\pi\left(\frac{J_{LL}^2\rho_L^2 + J_{RR}^2\rho_R^2}{2} + J_{LR}^2\rho_L\rho_R\right)T\varphi\left(\frac{\Delta}{T}\right), \tag{27}$$

$$\frac{1}{T_2^{\rm eq}} = \frac{1}{2T_1^{\rm eq}} + 4\pi\left(\frac{J_{LL}^2\rho_L^2 + J_{RR}^2\rho_R^2}{2} + J_{LR}^2\rho_L\rho_R\right)T. \quad (28)$$

It is clear that the contribution of non-spin-flip cotunneling to the pure decoherence rate, the second term in Eq. (28), is proportional to temperature, while spin-flip cotunneling leads to a somewhat complicated temperature dependence, $\Delta\coth(\Delta/2T)$. This difference can also be ascribed to energy exchange in the dissipation process. It is worth noting that without transport ($V\to 0$), dissipation (relaxation and decoherence) is due solely to quantum thermal fluctuations (thermal noise). Furthermore, if the external magnetic field is quenched, the thermal fluctuations are purely non-energy-consuming:

$$\tau_{\rm th} = \frac{1}{T_1^{\rm eq}} = \frac{1}{T_2^{\rm eq}} = 8\pi\left(\frac{J_{LL}^2\rho_L^2 + J_{RR}^2\rho_R^2}{2} + J_{LR}^2\rho_L\rho_R\right)T, \quad (29)$$

indicating that the dissipation is totally determined by thermodynamics (temperature) of the reservoirs.

For illustrative purposes, we exhibit in Fig. 1 the dependences of the relaxation rate and the dephasing rate on magnetic field (a,b) and on bias-voltage (c,d) for given temperatures. At relatively low temperatures, $T/V=0.01$ (or $T/\Delta=0.01$), these rates show linear increase with respect to bias-voltage V (magnetic field Δ) with the rates of increase depending on the relative magnitudes of V and Δ. Interestingly, dephasing, $1/T_1$, is independent of V for $V<\Delta$, as shown in Fig. 1(c). This comes about because the hyperbolic cotangent functions behave as $\varphi((\Delta+V)/T)+\varphi((\Delta-V)/T)\to|\Delta+V|+|\Delta-V|$ in the limit $T\to 0$. As expected, rising temperature smears out the low-temperature structures in these rates [Figs. 1(b) and 1(c)]. Finally, the temperature dependences of the two rates are summarized in Fig. 2.

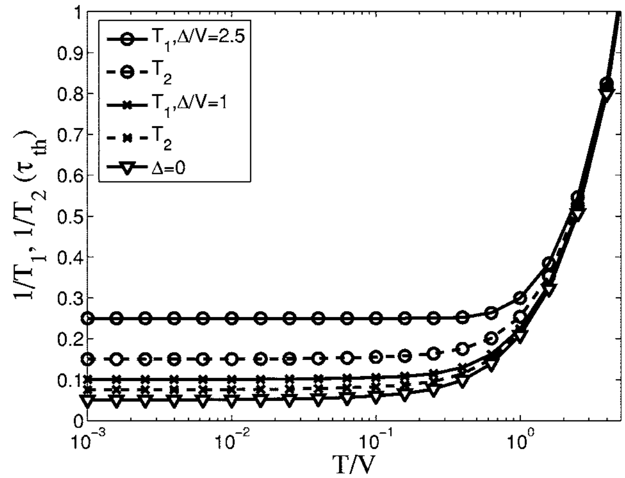

FIG. 2. The cotunneling-induced spin relaxation rate (T_1^{-1}) and decoherence rate (T_2^{-1}) as functions of temperature for $\Delta/V=2.5$, 1.0, and 0. Other parameters are the same as in Fig. 1.

The magnetization of the QD, defined as $M=S^z$, is readily obtained using the steady solution of Eq. (22) as

$$M(\Delta,V) = S_\infty^z = \frac{R(\Delta)}{2C(\Delta)} = \frac{\left(\frac{J_{LL}^2\rho_L^2 + J_{RR}^2\rho_R^2}{2} + J_{LR}^2\rho_L\rho_R\right)\frac{\Delta}{T}}{(J_{LL}^2\rho_L^2 + J_{RR}^2\rho_R^2)\varphi\left(\frac{\Delta}{T}\right) + J_{LR}^2\rho_L\rho_R\left[\varphi\left(\frac{\Delta+V}{T}\right) + \varphi\left(\frac{\Delta-V}{T}\right)\right]}, \quad (30)$$

which is identical to previous theoretical result.[8,9] In absence of bias-voltage, $V=0$, it reduces to the equilibrium expression $M(\Delta,0)=\frac{1}{2}\tanh(\Delta/2T)$.

V. NONLINEAR TUNNELING CURRENT

The calculation of steady state tunneling charge current, I^c, measuring the charge flow from left lead to right lead, is based on the equation of motion for the charge density $N_L=\Sigma_\sigma N_{L\sigma}=\Sigma_{\mathbf{k}} c_{L\mathbf{k}\sigma}^\dagger c_{L\mathbf{k}\sigma}$ in the left lead, which is a sum of both spin-up and spin-down electrons flows, $I^c=I_{L\uparrow}+I_{L\downarrow}$,

$$\begin{aligned} I_{L\uparrow} &= -\langle \dot{N}_{L\uparrow}\rangle \\ &= i\langle[N_{L\uparrow},H]\rangle \\ &= i\langle (Q_{LR}^{z\uparrow\uparrow} - Q_{RL}^{z\uparrow\uparrow})S^z - (Q_{LL}^- + Q_{RL}^-)S^+ + (Q_{LL}^+ + Q_{LR}^+)S^-\rangle, \end{aligned} \quad (31)$$

$$\begin{aligned} I_{L\downarrow} &= -\langle \dot{N}_{L\downarrow}\rangle \\ &= i\langle[N_{L\downarrow},H]\rangle \\ &= i\langle (Q_{RL}^{z\downarrow\downarrow} - Q_{LR}^{z\downarrow\downarrow})S^z + (Q_{LL}^- + Q_{LR}^-)S^+ - (Q_{LL}^+ + Q_{RL}^+)S^-\rangle, \end{aligned} \quad (32)$$

with the definition $Q_{\eta\eta'}^{z\sigma\sigma}=J_{\eta\eta'}\Sigma_{\mathbf{k},\mathbf{k}'}c_{\eta\mathbf{k}\sigma}^\dagger c_{\eta'\mathbf{k}'\sigma}$. Using the same procedure described in the preceding section and Appendix A, and employing the various response and correlation functions of the free reservoir variables determined in Appendix B, we have derived analytic expressions for the spin-resolved current, $I_{L\sigma}$. For example, the spin-up current, $I_{L\uparrow}$, takes the form

$$I_{L\uparrow}=\frac{1}{2}\int_{-\infty}^{t}d\tau[R^{zz\uparrow\uparrow}_{LR,RL}(\tau)-R^{zz\uparrow\uparrow}_{RL,LR}(\tau)]$$
$$-\int_{-\infty}^{t}d\tau\{e^{-i\Delta\tau}[R^{-+}_{LL,LL}(\tau)+R^{-+}_{RL,LR}(\tau)]-e^{i\Delta\tau}[R^{+-}_{LL,LL}(\tau)$$
$$+R^{+-}_{LR,RL}]\}-2S^{z}\int_{-\infty}^{t}d\tau\{e^{-i\Delta\tau}[C^{-+}_{LL,LL}(\tau)+C^{-+}_{RL,LR}(\tau)]$$
$$+e^{i\Delta\tau}[C^{+-}_{LL,LL}(\tau)+C^{+-}_{LR,RL}(\tau)]\}. \quad (33)$$

Using Eqs. (B4), (B5), and (B9)–(B11), and then making the replacement $\int_{-\infty}^{t}d\tau\Rightarrow\int_{-\infty}^{\infty}d\tau$, we finally arrived at an explicit result for $I_{L\uparrow}$ as a function of temperature and bias-voltage [after performing the ϵ integrals of Eqs. (B16) and (B17)],

$$I_{L\uparrow}=\frac{3\pi}{2}J^{2}_{LR}\rho_{L}\rho_{R}V+\pi\Delta(J^{2}_{LL}\rho^{2}_{L}+J^{2}_{LR}\rho_{L}\rho_{R})$$
$$-2\pi T\left[J^{2}_{LL}\rho^{2}_{L}\varphi\left(\frac{\Delta}{T}\right)+J^{2}_{LR}\rho_{L}\rho_{R}\varphi\left(\frac{\Delta+V}{T}\right)\right]S^{z}. \quad (34)$$

Similarly, the current from spin-down electrons, $I_{L\downarrow}$, is given by

$$I_{L\downarrow}=\frac{3\pi}{2}J^{2}_{LR}\rho_{L}\rho_{R}V-\pi\Delta(J^{2}_{LL}\rho^{2}_{L}+J^{2}_{LR}\rho_{L}\rho_{R})$$
$$+2\pi T\left[J^{2}_{LL}\rho^{2}_{L}\varphi\left(\frac{\Delta}{T}\right)+J^{2}_{LR}\rho_{L}\rho_{R}\varphi\left(\frac{\Delta-V}{T}\right)\right]S^{z}. \quad (35)$$

The total charge current, I^{c}, is thus

$$I^{c}=\pi J^{2}_{LR}\rho_{L}\rho_{R}\left\{3V+2S^{z}T\left[\varphi\left(\frac{\Delta-V}{T}\right)-\varphi\left(\frac{\Delta+V}{T}\right)\right]\right\}. \quad (36)$$

It is worth noting that the cotunneling current, Eq. (36), is just proportional to second order in the exchange coupling constant, J_{LR}, and higher-order contributions are all neglected. This is because we employ the approximation formula, Eq. (A2), to derive non-Markovian quantum dynamic equations and the current, leading to the absence of the characteristic logarithmic divergence term in current. Thus, the present approach *cannot* be applied to describe strong Kondo correlations, but can be used to study the ordinary cotunneling process in the weak tunnel-coupling limit.

Due to the fact that spin-up electrons are coupled with spin-down electrons via the spin-flip processes in this model, there is an imbalance between the spin-up current and spin-down current, i.e., there is a net spin current, I^{s}, with respect to the left lead, defined as

$$I^{s}=I_{L\uparrow}-I_{L\downarrow}$$
$$=2\pi\Delta(J^{2}_{LL}\rho^{2}_{L}+J^{2}_{LR}\rho_{L}\rho_{R})-2\pi T\left\{2J^{2}_{LL}\rho^{2}_{L}\varphi\left(\frac{\Delta}{T}\right)\right.$$
$$\left.+J^{2}_{LR}\rho_{L}\rho_{R}\left[\varphi\left(\frac{\Delta-V}{T}\right)+\varphi\left(\frac{\Delta+V}{T}\right)\right]\right\}S^{z}. \quad (37)$$

To better understand these formulas and the physical perspective involved, we elaborate the physical picture of cotunneling processes through a QD in a finite magnetic field. When the electronic levels of the QD, $\epsilon_{d\sigma}$, are far below the chemical potentials of the two leads, i.e., $\epsilon_{d\sigma}\ll\mu_{L(R)}$, the first-order tunneling process, sequential tunneling, vanishes. However, higher-order tunneling processes, *cotunneling*, are active and dominate the quantum transport. In the strong Coulomb blockade regime, the QD is always *singly* occupied by an electron because of the deep electronic energy, $\epsilon_{d\sigma}$, and the extremely strong charging energy, $U\to\infty$, involved when an additional, excess electron attempts to enter the QD, i.e., $\epsilon_{d\sigma}+U\gg\mu_{L(R)}$. This steady occupation means that *no charge fluctuation* takes place, provided that the applied bias-voltage is not strong enough to force the chemical potential in one of leads below the QD level, $\epsilon_{d\sigma}$, so as to drive the transport into the sequential tunneling regime. Therefore, in nonequilibrium conditions, a cotunneling event consists of two single-particle tunneling processes, ① and ② which can take place in sequence as follows: event ①, an electron inside the QD with spin σ will at first tunnel out to a lead L or R, inducing a virtual empty state in the dot, but this is immediately followed by a second single-particle tunneling event ② in which an electron in one of the leads is injected into the QD with the same spin σ (spin-conserving elastic cotunneling) or with the opposite spin (spin-flip inelastic cotunneling). *The two tunneling events occur via a virtual empty-dot state in a very short time interval to insure coherence.* Importantly, spin-flip cotunneling provides a mechanism for the spin orientation of the QD to be changed, which is fully quantum phenomenology. Moreover, it stimulates intrinsic *spin fluctuation*, which is a fundamental concept in the context of Kondo physics. Obviously, there is a total of 16 different cotunneling events allowed in this strong Coulomb blockade system, which are schematically shown in Fig. 3. We can classify them in four different categories/types.

Type-I cotunneling involves only one lead and equal spin orientations in two successive single-particle tunneling events. Figures 3(a)–3(d) depict such trivial cotunneling events, in which an electron with spin σ exits the QD to the lead η and subsequently an electron with the same spin in the same lead (probably not the same electron) transfers to the QD. Obviously, these events make no contribution to the current. In contrast, type-II cotunneling experiences a spin-flip process as shown in Figs. 3(e)–3(h), in which the final spin state in the QD is opposite to its initial state. Of course, only events Figs. 3(e) and 3(f) relate to the currents in the left lead, and involve the terms $\mp Q^{-}_{LL}S^{+}$ and $\pm Q^{+}_{LL}S^{-}$ in Eqs. (31) and (32). However, the spin-up and spin-down electrons flow in opposite directions and contribute to the currents with equal magnitudes. As a result, no charge current occurs in this type cotunneling but spin current does emerge. Furthermore, because only one lead is involved, the contributions of events Figs. 3(e) and 3(f) are naturally independent of the bias-voltage and are only dependent on the magnetic field, involving the terms $\pm\pi\Delta J^{2}_{LL}\rho^{2}_{L}\mp2\pi TJ^{2}_{LL}\rho^{2}_{L}\varphi(\Delta/T)S^{z}$ in spin-up and spin-down currents with $\pm\to+,-$, respectively. Figures 3(i)–3(l) represent all type-III cotunneling processes. This kind of cotunneling describes an equivalent spin-

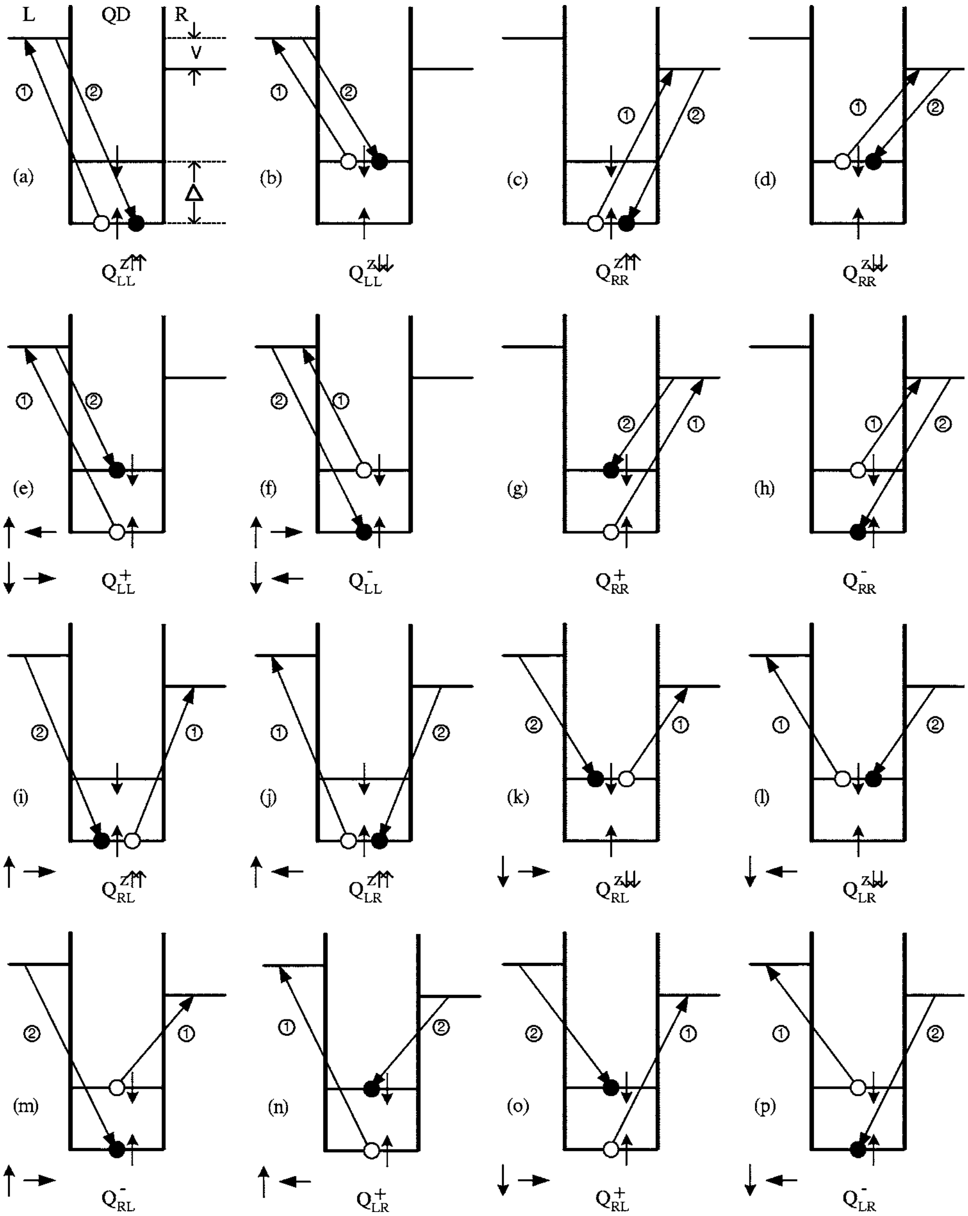

FIG. 3. Schematic description of all 16 cotunneling processes (a)–(p) through a QD subject to Zeeman splitting energy Δ between the spin-up and -down electronic states, in the strong Coulomb blockade regime. A finite bias-voltage V is applied between the two leads, L and R. An open circle inside the QD stands for the initial occupied electron state before tunneling events, while the solid circle denotes the final state occupied by an electron after the cotunneling processes. The single-particle tunneling event ① takes place first and then is followed by the tunneling event ②, which together comprise the entire cotunneling process. The reservoir variable below each of the figures denotes the corresponding physical process occurring in the reservoir. The arrow beneath the left lead in each of the figures denotes the flow direction of the spin-up and/or spin-down electron with respect to the left lead.

conservative tunneling process, in which an electron is transferred from one lead to another lead via the QD without spin exchange. The corresponding terms in Eqs. (31) and (32) are $\pm Q^{z\uparrow\uparrow(\downarrow\downarrow)}_{LR(RL)}S^z$. Moreover, we observe that (1) the spin-up events Figs. 3(i) and 3(j) and the spin-down events Figs. 3(k) and 3(l) yield currents having not only the same directions, but they also have equal magnitudes; and (2) these spin-up (down) events involve only the difference of chemical potentials, leading to a contribution proportional to the bias-voltage, $\frac{3}{2}\pi J^2_{LR}\rho_L\rho_R V$. This observation reveals that type-III cotunneling excludes the possibility of spin current, but it does provide a linearly bias-voltage-dependent term in the charge current, Eq. (36). Finally, type-IV cotunneling comprises the four electron-transferring tunneling events accompanied by a spin-flip process as exhibited in Figs. 3(m)–3(p). They produce the terms $-Q^-_{RL}S^+$, $+Q^+_{LR}S^-$ in Eq. (31) and $Q^-_{LR}S^+, -Q^+_{RL}S^-$ in Eq. (32). Differing from type-III cotunneling, we find that in type-IV cotunneling the spin-up (down) events Figs. 3(m) and 3(n) [or Figs. 3(o) and 3(p)] involve both the voltage change and spin flip, and the corresponding contributions to current are dependent on both V and Δ: $\pm[\Delta-2S^zT\varphi(\Delta\pm V/T)]\pi J^2_{LR}\rho_L\rho_R$. Type-IV cotunneling produces both spin and charge currents. In sum, the mechanism for creating spin current stems solely from inelastic spin-flip cotunneling processes (type-II and -IV), while the electron-transferring elastic and inelastic cotunneling processes (type-III and IV) are responsible for producing charge current.

Substituting the steady-state solution, Eq. (30), into the charge current, Eq. (36), and the spin current, Eq. (37), we readily find that (1) both the charge current and the spin current are zero if $V=0$; (2) the resulting spin current is nonzero in nonequilibrium conditions, $V\neq 0$, if and only if two conditions are satisfied: $J_{LL}\neq J_{RR}$, i.e., the asymmetrical Kondo coupling case, and there is a nonvanishing magnetic field, $\Delta\neq 0$; (3) the spin current is an even function of the applied bias-voltage, V, indicating that the sign of spin cur-

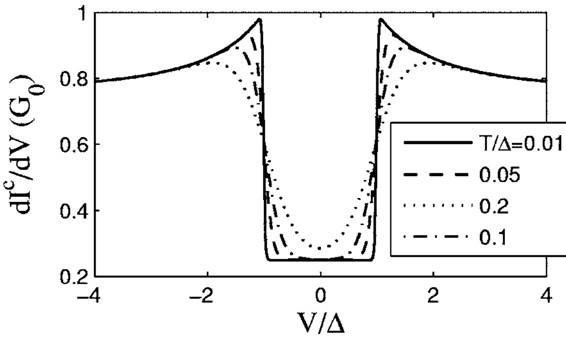

FIG. 4. The calculated differential conductance dI^c/dV vs bias-voltage V/Δ for several temperatures at nonzero magnetic field in units of $G_0=4\pi J_{LR}^2\rho_L\rho_R$ (linear conductance at zero magnetic field). Other parameters are the same as in Fig. 1.

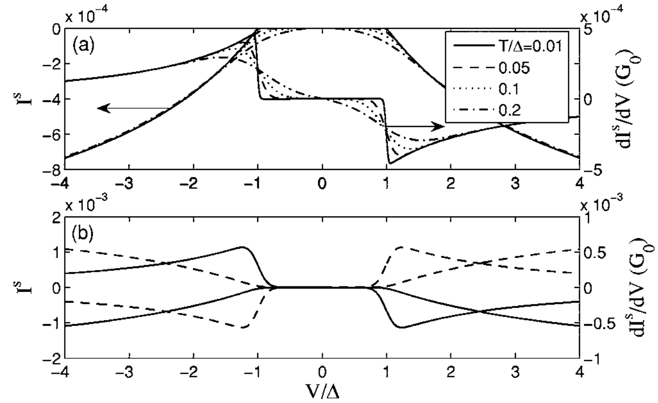

FIG. 5. The calculated spin current, I^s, and its differential conductance, dI^s/dV, as functions of bias-voltage V/Δ, at nonzero magnetic field. (a) Exhibits results for several temperatures and $J_{RR}/J_{LL}=4.0$, $J_{LL}=0.02$. (b) Plots the results for $J_{RR}/J_{LL}=5.0$ ($J_{LL}=0.02$) as solid lines, and for $J_{LL}/J_{RR}=5.0$ ($J_{RR}=0.02$) as dashed lines.

rent is not related to the direction of the bias-voltage; whereas charge current, Eq. (36), is an odd function of the bias-voltage and will change its sign when bias-voltage is applied in the opposite direction; (4) the magnetic-field-related spin current changes its sign when the direction of the applied magnetic field is reversed (odd function), while the charge current is an even function of Δ, because it measures the results of the total charge flow irrespective of the spin orientation. The sign property of the spin current was also pointed out in previous study.[19]

As an illustration, we plot the bias-voltage-dependent differential conductance, dI^c/dV, in Fig. 4. The differential conductance shows a characteristic jump at $V=\pm\Delta$, which is the signature of the Kondo effect in the presence of an external magnetic field. Mathematically, this feature comes from the hyperbolic cotangent function in the current formula, Eq. (36). From a physical point of view, this splitting can be qualitatively understood from the following consideration: a small bias-voltage, $|V|<\Delta$, cannot provide enough energy to spur the spin-flip cotunneling process that is an energy-consuming event in the case of nonzero magnetic field; however, when $|V|\geqslant\Delta$, the spin-flip cotunneling process is energetically activated, thus an additional channel is opened for electron transport. Moreover, the effect of temperature is to smear and reduce the peak.

We also exhibit the resulting spin current, I^s, and its differential conductance, defined as dI^s/dV, as functions of bias-voltage V/Δ in Fig. 5. We observe that at low temperatures, $T/\Delta=0.01$ and 0.05 in Fig. 5(a), the calculated spin currents are nearly zero in the small bias-voltage region, $|V|<\Delta$, notwithstanding $J_{LL}/J_{RR}=4.0$ and $\Delta\neq 0$. Analogous to the peak splitting of the differential conductance shown in Fig. 4, the low-temperature vanishing of spin current is also due to the fact that spin-flip scattering is energetically inaccessible in the case of small bias-voltage. This vanishing produces a "window" of zero differential conductance for spin current. Nevertheless, at higher temperatures, thermal fluctuation provides an additional possibility to flip spin in the tunneling processes, leading to a slow increase of the spin current and the gradual disappearance of the zero "window" in dI^s/dV. In Fig. 5(b), we show that the sign of the spin current is also determined by the relative magnitudes of J_{LL} and J_{RR}.

VI. CONCLUSIONS

In this paper, we have systematically examined nonequilibrium inelastic cotunneling through a single spin (QD) subject to a finite magnetic field in the strong Coulomb blockade regime, in the weak tunnel coupling limit. For this purpose, we introduced the Kondo Hamiltonian to model cotunneling in the QD and employed a generic Heisenberg-Langevin equation approach to establish a set of quantum Bloch-type dynamical equations describing inelastic cotunneling phenomenology.

In our formulation, the operators of the localized spin and the reservoirs were first determined formally by integration of their Heisenberg equations of motion, exactly to all orders in the tunnel coupling constants. Next, under the assumption that the time scale of the decay processes is much slower than that of free evolution, we expressed the time-dependent operators involved in the integrands of these equations of motion approximately in terms of their free evolution. Third, these equations of motion were expanded in powers of the tunnel-coupling constants to second order; this approximation is physically valid in the weak tunnel-coupling limit. On the basis of these consideration, jointly with normal ordering, we developed the Bloch-type equations expressed explicitly and compactly in terms of the response and the correlation functions of the free reservoir variables, which facilitated our theoretical examination of relaxation and decoherence in the localized spin induced by the "environment."

In the problem at hand, dissipation of the QD spin stems from tunnel-coupling of the QD to two leads by cotunneling mechanisms. Based on our derived Bloch equations, we obtained explicit analytical expressions for the corresponding relaxation and decoherence rates at arbitrary bias-voltage and temperature. We found that relaxation results exclusively from spin-flip cotunneling processes alone, whereas both spin-flip and non-spin-flip cotunneling events contribute to decoherence. In this analysis, we carried out systematic examinations of the relaxation rate and the decoherence rate as functions of bias-voltage, external magnetic field, and temperature. Our formulation also facilitated the derivation of an analytic expression for the nonequilibrium magnetization that is found to match that of earlier theories.

Employing this approach, we also derived closed-form expressions for the spin-resolved currents, which facilitated our calculation of both the charge current and the spin current. Furthermore, we classified and examined all possible cotunneling processes occurring in the strong Coulomb interaction QD (16 events), and categorized them in four distinct types. In this, we found that (1) type-I cotunneling make no contribution to current; (2) spin-flip processes, types-II and IV cotunneling, drive the spin current; (3) the electron-transferring processes, types-III (non-spin-flip) and IV (spin-flip) cotunneling, produce charge current; and (4) we also determined formulas for their respective contributions to the charge and spin currents. Our numerical calculations exhibit splitting of the zero-bias-voltage peak in the differential conductance for charge current in a finite magnetic field, which is a typical signature of the Kondo effect, and a wide "window" of zero differential conductance for spin current about zero-bias-voltage. With insight gained from our specific analyses, we can ascribe these low-temperature transport characteristics to the fact that *inelastic* spin-flip cotunneling is energetically active only for sufficiently strong applied bias-voltage, $V \geqslant \Delta$. We have also shown that spin current, unlike charge current, is an even function of the applied bias-voltage, and its direction depends on the orientation of the ambient magnetic field and asymmetry of the Kondo coupling constants to the left and the right leads.

ACKNOWLEDGMENTS

This work was supported by the Department of Defense through the DURINT Program administered by the U.S. Army Research Office, DAAD Grant No.19-01-1-0592.

APPENDIX A: DERIVATION OF EQS. (10a), (12), AND (13)

In this Appendix, we first prove Eqs. (10a) and (10b). Consider $Q^z_{\eta\eta'}$ for example. Substituting Eqs. (9a) and (9b) into the definition of Eq. (2b), we have

$$
\begin{aligned}
Q^z_{\eta\eta'}(t) &= J_{\eta\eta'}\sum_{\mathbf{k},\mathbf{k}'}[c^{o\dagger}_{\eta\mathbf{k}\uparrow}(t)+c^{i\dagger}_{\eta\mathbf{k}\uparrow}(t)][c^{o}_{\eta'\mathbf{k}'\uparrow}(t)+c^{i}_{\eta'\mathbf{k}'\uparrow}(t)] \\
&\quad - J_{\eta\eta'}\sum_{\mathbf{k},\mathbf{k}'}(\uparrow\to\downarrow) \\
&= Q^{zo}_{\eta\eta'}(t) - iJ_{\eta\eta'}\sum_{\mathbf{k},\mathbf{k}'}\int_0^t dt' \\
&\quad \times\{c^{o\dagger}_{\eta\mathbf{k}\uparrow}(t){:}[c^{o}_{\eta'\mathbf{k}'\uparrow}(t),H^o_{\mathrm{I}}(t')]_- \\
&\quad - {:}[c^{o\dagger}_{\eta\mathbf{k}\uparrow}(t),H^o_{\mathrm{I}}(t')]_-{:}c^{o}_{\eta'\mathbf{k}'\uparrow}(t)\} + iJ_{\eta\eta'}\sum_{\mathbf{k},\mathbf{k}'}(\uparrow\to\downarrow) \\
&= Q^{zo}_{\eta\eta'}(t) - iJ_{\eta\eta'}\sum_{\mathbf{k},\mathbf{k}'}\theta(\tau)\int_0^\infty dt' \\
&\quad \times {:}[c^{o\dagger}_{\eta\mathbf{k}\uparrow}(t)c^{o}_{\eta'\mathbf{k}'\uparrow}(t),H^o_{\mathrm{I}}(t')]_-{:} + iJ_{\eta\eta'}\sum_{\mathbf{k},\mathbf{k}'}(\uparrow\to\downarrow) \\
&= Q^{zo}_{\eta\eta'}(t) - i\theta(\tau)\int_{-\infty}^t d\tau{:}[Q^{zo}_{\eta\eta'}(t),H^o_{\mathrm{I}}(t')]_-{:}.
\end{aligned} \tag{A1}
$$

In the second stage of Eq. (A1), we neglect terms of the form, $c^{i\dagger}_{\eta\mathbf{k}\sigma}c^{i}_{\eta'\mathbf{k}'\sigma'}$, since they are of second order in the coupling constant, $O(J^2)$, yielding a third-order contribution to $Q^z_{\eta\eta'}$ with respect to J.

To derive Eqs. (12) and (13), we consider the normally ordered product of the reservoir and spin operators, ${:}Q^aS^b{:}$,

$$
\begin{aligned}
{:}Q^a(t)S^b(t) &\coloneqq {:}[Q^a_o(t)+Q^a_i(t)][S^b_o(t)+S^b_i(t)]{:} \\
&= {:}Q^a_o(t)S^b_o(t){:} \\
&\quad - i\theta(\tau)\int_{-\infty}^t d\tau\{{:}Q^a_o(t){:}[S^b_o(t),H^o_{\mathrm{I}}(t')]_-{:} \\
&\quad + {:}[Q^a_o(t),H^o_{\mathrm{I}}(t')]_-{:}S^b_o(t){:}\}.
\end{aligned} \tag{A2}
$$

Once again, we neglect the term $Q^a_iS^b_i$ as it is proportional to $O(J^3)$. The first term in Eq. (A2) involves only the free reservoir variables and the decoupled single spin. The other interaction terms (we designate the operator expressions in the integrand as $\mathcal{I}$) arise from tunneling reaction, upon which we focus in the following derivation. Using the compact definition of the interaction Hamiltonian, Eq. (2a), $H_{\mathrm{I}}=\sum_{c\in\{z,+,-\}}Q^cF_{Q^c}$, we have

$$
\begin{aligned}
\mathcal{I} &= \sum_c\{{:}Q^a_o(t)Q^c_o(t')[S^b_o(t),F^o_{Q^c}(t')]_- \\
&\quad + [Q^a_o(t),Q^c_o(t')]_-F^o_{Q^c}(t')S^b_o(t){:}\} \\
&= \sum_c\Bigg\{{:}\frac{1}{2}[Q^a_o(t),Q^c_o(t')]_+[S^b_o(t),F^o_{Q^c}(t')]_- \\
&\quad + \frac{1}{2}[Q^a_o(t),Q^c_o(t')]_-[S^b_o(t),F^o_{Q^c}(t')]_- \\
&\quad + [Q^a_o(t),Q^c_o(t')]_-F^o_{Q^c}(t')S^b_o(t){:}\Bigg\} \\
&= \frac{1}{2}\sum_c\{{:}[Q^a_o(t),Q^c_o(t')]_+[S^b_o(t),F^o_{Q^c}(t')]_- \\
&\quad + [Q^a_o(t),Q^c_o(t')]_-[S^b_o(t),F^o_{Q^c}(t')]_+{:}\}.
\end{aligned} \tag{A3}
$$

Therefore, the full normal-ordered operator product (${:}Q^aS^b{:}$) is written as the sum of a zero-order term and a term of second-order in the coupling constant J, having the compact form:

$$
\begin{aligned}
{:}Q^a(t)S^b(t) \coloneqq {:}Q^a_o(t)S^b_o(t){:} - i\int_{-\infty}^t d\tau\sum_c\{{:}\hat{C}^{ac}(t,t') \\
\times[S^b_o(t),F^o_{Q^c}(t')]_- + \hat{R}^{ac}(t,t')[S^b_o(t),F^o_{Q^c}(t')]_+{:}\},
\end{aligned} \tag{A4}
$$

with the definitions

$$\hat{R}^{ac}(t,t') = \tfrac{1}{2}\theta(\tau)[Q_o^a(t), Q_o^c(t')]_-, \tag{A5}$$

$$\hat{C}^{ac}(t,t') = \tfrac{1}{2}\theta(\tau)[Q_o^a(t), Q_o^c(t')]_+. \tag{A6}$$

The reservoir equilibrium ensemble averages of $\hat{R}^{ac}(t,t')$ and $\hat{C}^{ac}(t,t')$ are just the response function $R^{ac}(t,t')$ and the correlation function $C^{ac}(t,t')$ defined in Eqs. (14) and (15), respectively.

In the spin operator equations of motion, these zero-order terms contribute quantum fluctuations associated with the reservoir fields, as well as quantum effects pertaining to the intrinsic character of the reservoirs (for example, superconducting or ferromagnetic leads). In any event, we take the ensemble average of each equation of motion separately in regard to the electron ensembles of the reservoirs and in regard to the quantum spin states. Thus, the normally ordered operator products factorize in the averaging procedure. Considering that we take no account of quantum fluctuations in the present paper and only normal leads are connected to the single spin, the zero-order terms make no contribution to the quantum Bloch equations. Moreover, only $\langle Q_o^{\pm}(t)Q_o^{\mp}(t')\rangle_e$ and $\langle Q_o^z(t)Q_o^z(t')\rangle_e$ are nonzero for normal leads (see Appendix B). Combining all the above results, we obtain Eqs. (12) and (13).

APPENDIX B: RESPONSE AND CORRELATION FUNCTIONS OF THE RESERVOIRS

To obtain explicit expressions for the Bloch equations and the current, we need to determine the various correlation and response functions $C(R)^{ab}_{\eta_1\eta_2,\eta_3\eta_4}(t,t')$ of the free reservoir variables, which are defined as

$$C(R)^{ab}_{\eta_1\eta_2,\eta_3\eta_4}(t,t') = \frac{1}{2}\theta(\tau)\langle[Q^{ao}_{\eta_1\eta_2}(t), Q^{bo}_{\eta_3\eta_4}(t')]_\pm\rangle. \tag{B1}$$

In the following, we drop all super(sub)scripts, "o," bearing in mind that all operators are free reservoir operators. In our calculations, we assume that (i) the leads have a flat density of states ρ_η for both spin orientations, so we can make the replacement

$$\sum_{\mathbf{k}}(\cdots) \to \rho_\eta \int d\epsilon(\cdots); \tag{B2}$$

(ii) the normal leads are in the respective bias-voltage-driven local equilibrium states described by

$$f_\eta(\epsilon) = [1 + e^{(\epsilon-\mu_\eta)/T}]^{-1}, \tag{B3}$$

with temperature T and chemical potential μ_η; and (iii) the time evolution of free reservoir operators is governed by Eq. (8c). According to Wick's theorem and properties (i) and (ii), it is easy to see that only the functions $C(R)^{zz}_{\eta\eta',\eta'\eta}$, $C(R)^{+-}_{\eta\eta',\eta'\eta}$, and $C(R)^{-+}_{\eta\eta',\eta'\eta}$ are nonzero. We calculate them individually.

(1) $C(R)^{zz}(t,t') = \Sigma_{\eta,\eta'} C(R)^{zz}_{\eta\eta',\eta'\eta}(t,t')$:

$$\begin{aligned}
C(R)^{zz\uparrow\uparrow}_{LR,RL}(t,t') &= \frac{1}{2}\theta(\tau)\langle[Q^{z\uparrow\uparrow}_{LR}(t), Q^{z\uparrow\uparrow}_{RL}(t')]_\pm\rangle_e \\
&= \frac{1}{2}\theta(\tau)J^2_{LR}\sum_{\mathbf{k},\mathbf{k}',\mathbf{q},\mathbf{q}'}\langle[c^\dagger_{L\mathbf{k}\uparrow}(t)c^\dagger_{R\mathbf{k}'\uparrow}(t), c^\dagger_{R\mathbf{q}\uparrow}(t')c^\dagger_{L\mathbf{q}'\uparrow}(t')]_\pm\rangle_e \\
&= \frac{1}{2}\theta(\tau)J^2_{LR}\sum_{\mathbf{k},\mathbf{k}',\mathbf{q},\mathbf{q}'} e^{i(\epsilon_{L\mathbf{q}'}-\epsilon_{R\mathbf{q}})\tau} \\
&\quad\times[\langle c^\dagger_{L\mathbf{k}\uparrow}(t)c^\dagger_{L\mathbf{q}'\uparrow}(t)\rangle_e\langle c^\dagger_{R\mathbf{k}'\uparrow}(t)c^\dagger_{R\mathbf{q}\uparrow}(t)\rangle_e \\
&\quad\pm\langle c^\dagger_{R\mathbf{q}\uparrow}(t)c^\dagger_{R\mathbf{k}'\uparrow}(t)\rangle_e\langle c^\dagger_{L\mathbf{q}'\uparrow}(t)c^\dagger_{L\mathbf{k}\uparrow}(t)\rangle_e] \\
&= \frac{1}{2}\theta(\tau)J^2_{LR}\rho_L\rho_R\int d\epsilon d\epsilon' e^{i(\epsilon-\epsilon')\tau} \\
&\quad\times\{f_L(\epsilon)[1-f_R(\epsilon')]\pm f_R(\epsilon')[1-f_L(\epsilon)]\}.
\end{aligned} \tag{B4}$$

Exchanging the roles of R and L, $L\leftrightarrow R$, $C(R)^{zz\uparrow\uparrow}_{RL,LR}(t,t')$ yields

$$\begin{aligned}
C(R)^{zz\uparrow\uparrow}_{RL,LR}(t,t') &= \frac{1}{2}\theta(\tau)J^2_{LR}\rho_L\rho_R\int d\epsilon d\epsilon' e^{-i(\epsilon-\epsilon')\tau} \\
&\quad\times\{f_R(\epsilon')[1-f_L(\epsilon)]\pm f_L(\epsilon)[1-f_R(\epsilon')]\}.
\end{aligned} \tag{B5}$$

As we take the leads to be normal metals /(semiconductors), we have

$$\begin{aligned}
C(R)^{zz\downarrow\downarrow}_{LR,RL}(t,t') &= \frac{1}{2}\theta(\tau)\langle[Q^{z\downarrow\downarrow}_{LR}(t), Q^{z\downarrow\downarrow}_{RL}(t')]_\pm\rangle_e \\
&= C(R)^{zz\uparrow\uparrow}_{LR,RL}(t,t'),
\end{aligned}$$

$$C(R)^{zz\downarrow\downarrow}_{RL,LR}(t,t') = C(R)^{zz\uparrow\uparrow}_{RL,LR}(t,t'),$$

$$C(R)^{zz\uparrow\downarrow}_{LR,RL}(t,t') = \frac{1}{2}\theta(\tau)\langle[Q^{z\uparrow\uparrow}_{LR}(t), Q^{z\downarrow\downarrow}_{RL}(t')]_\pm\rangle_e = 0,$$

$$C(R)^{zz\uparrow\downarrow}_{LR,RL}(t,t') = 0.$$

Furthermore,

$$\begin{aligned}
C(R)^{zz}_{LL,LL}(t,t') &= \frac{1}{2}\theta(\tau)\langle[Q^z_{LL}(t), Q^z_{LL}(t')]_\pm\rangle_e \\
&= \theta(\tau)J^2_{LL}\rho^2_L\int d\epsilon d\epsilon' e^{i(\epsilon-\epsilon')\tau} \\
&\quad\times\{f_L(\epsilon)[1-f_L(\epsilon')]\pm f_L(\epsilon')[1-f_L(\epsilon)]\},
\end{aligned} \tag{B6}$$

$$C(R)^{zz}_{RR,RR}(t,t') = \frac{1}{2}\theta(\tau)\langle[Q^z_{RR}(t),Q^z_{RR}(t')]_\pm\rangle_e$$
$$= \theta(\tau)J^2_{RR}\rho^2_R \int d\epsilon d\epsilon' e^{i(\epsilon-\epsilon')\tau}$$
$$\times \{f_R(\epsilon)[1-f_R(\epsilon')] \pm f_R(\epsilon')[1-f_R(\epsilon)]\}. \quad \text{(B7)}$$

Finally, $C(R)^{zz}(t,t')$ are functions only of the time difference τ and take the form:

$$C(R)^{zz}(\tau) = \theta(\tau)\sum_\eta J^2_{\eta\eta}\rho^2_\eta \int d\epsilon d\epsilon' e^{i(\epsilon-\epsilon')\tau}$$
$$\times \{f_\eta(\epsilon)[1-f_\eta(\epsilon')] \pm f_\eta(\epsilon')[1-f_\eta(\epsilon)]\}$$
$$+ \theta(\tau)J^2_{LR}\rho_L\rho_R \int d\epsilon d\epsilon'[e^{i(\epsilon-\epsilon')\tau} \pm e^{-i(\epsilon-\epsilon')\tau}]$$
$$\times \{f_L(\epsilon)[1-f_R(\epsilon')] \pm f_R(\epsilon')[1-f_L(\epsilon)]\}. \quad \text{(B8)}$$

(2) $C(R)^{+-/-+}(t,t') = \Sigma_{\eta,\eta'} C(R)^{+-/-+}_{\eta\eta',\eta'\eta}(t,t')$:

$$C(R)^{+-/-+}_{LR,RL}(\tau) = \frac{1}{2}\theta(\tau)\langle[Q^{+/-}_{LR}(t),Q^{-/+}_{RL}(t')]_\pm\rangle_e$$
$$= \frac{1}{2}\theta(\tau)J^2_{LR}\rho_L\rho_R \int d\epsilon d\epsilon' e^{i(\epsilon-\epsilon')\tau}$$
$$\times \{f_L(\epsilon)[1-f_R(\epsilon')] \pm f_R(\epsilon')[1-f_L(\epsilon)]\}, \quad \text{(B9)}$$

$$C(R)^{+-/-+}_{RL,LR}(\tau) = \frac{1}{2}\theta(\tau)\langle[Q^{+/-}_{RL}(t),Q^{-/+}_{LR}(t')]_\pm\rangle_e$$
$$= \frac{1}{2}\theta(\tau)J^2_{LR}\rho_L\rho_R \int d\epsilon d\epsilon' e^{i(\epsilon-\epsilon')\tau}$$
$$\times \{f_R(\epsilon)[1-f_L(\epsilon')] \pm f_L(\epsilon')[1-f_R(\epsilon)]\}, \quad \text{(B10)}$$

$$C(R)^{+-/-+}_{\substack{LL,LL\\RR,RR}}(\tau) = \frac{1}{2}\theta(\tau)\left\langle\left[Q^{+/-}_{\substack{LL\\RR}}(t),Q^{-/+}_{\substack{LL\\RR}}(t')\right]_\pm\right\rangle_e$$
$$= \frac{1}{2}\theta(\tau)J^2_{LL/RR}\rho^2_{L/R} \int d\epsilon d\epsilon' e^{i(\epsilon-\epsilon')\tau}$$
$$\times \{f_{L/R}(\epsilon)[1-f_{L/R}(\epsilon')]$$
$$\pm f_{L/R}(\epsilon')[1-f_{L/R}(\epsilon)]\}. \quad \text{(B11)}$$

Moreover, we can easily obtain

$$C(R)^{+-}(\tau) = C(R)^{-+}(\tau) = \frac{1}{2}C(R)^{zz}(\tau). \quad \text{(B12)}$$

Therefore, in ω-Fourier space, the spectral function $C(\omega)$ defined in Eq. (17) is given by

$$C(\omega) = \frac{1}{2}\int_{-\infty}^{\infty} d\tau e^{i\omega\tau}C^{zz}(\tau)$$
$$= \frac{1}{2}\pi\sum_\eta J^2_{\eta\eta}\rho^2_\eta \int d\epsilon\{f_\eta(\epsilon)[1-f_\eta(\epsilon+\omega)]$$
$$+ f_\eta(\epsilon+\omega)[1-f_\eta(\epsilon)]\}$$
$$+ \frac{1}{2}\pi J^2_{LR}\rho_L\rho_R \int d\epsilon \sum_\eta \{f_\eta(\epsilon)[1-f_{\bar\eta}(\epsilon+\omega)]$$
$$+ f_\eta(\epsilon)[1-f_{\bar\eta}(\epsilon-\omega)]\}. \quad \text{(B13)}$$

Also, the imaginary part of the retarded susceptibility $R(\omega)$ defined in Eq. (16) is

$$R(\omega) = \frac{1}{2}\int_{-\infty}^{\infty} d\tau e^{i\omega\tau}R^{zz}(\tau)$$
$$= \frac{1}{2}\pi\sum_\eta J^2_{\eta\eta}\rho^2_\eta \int d\epsilon[f_\eta(\epsilon) - f_\eta(\epsilon+\omega)]$$
$$+ \frac{1}{2}\pi J^2_{LR}\rho_L\rho_R \int d\epsilon[f_R(\epsilon-\omega) - f_R(\epsilon+\omega)]. \quad \text{(B14)}$$

It is readily seen that

$$C(-\omega) = C(\omega), \quad R(-\omega) = -R(\omega). \quad \text{(B15)}$$

Using the formulas

$$\int d\epsilon[f_\eta(\epsilon+\omega) - f_{\eta'}(\epsilon)] = -(\mu_{\eta'} - \mu_\eta + \omega), \quad \text{(B16)}$$

and

$$\int d\epsilon f_\eta(\epsilon+\omega)[1-f_{\eta'}(\epsilon)] = \frac{\omega - \mu_\eta + \mu_{\eta'}}{e^{(\omega-\mu_\eta+\mu_{\eta'})/T} - 1}, \quad \text{(B17)}$$

we can perform the ϵ integrals in the large bandwidth limit, with the results

$$R(\omega) = \pi\left(\frac{J^2_{LL}\rho^2_L + J^2_{RR}\rho^2_R}{2} + J^2_{LR}\rho_L\rho_R\right)\omega, \quad \text{(B18)}$$

$$C(\omega) = \frac{\pi}{2}(J^2_{LL}\rho^2_L + J^2_{RR}\rho^2_R)T\varphi\left(\frac{\omega}{T}\right)$$
$$+ \frac{\pi}{2}J^2_{LR}\rho_L\rho_R T\left[\varphi\left(\frac{\omega+V}{T}\right) + \varphi\left(\frac{\omega-V}{T}\right)\right], \quad \text{(B19)}$$

where we have defined

$$\varphi(x) \equiv x\coth\left(\frac{x}{2}\right). \quad \text{(B20)}$$

It should be noted that when leads are in thermodynamic equilibrium, $\mu_L = \mu_R$, the spectral function $C(\omega)$ and the function $R(\omega)$ obey the fluctuation-dissipation theorem $C(\omega) = R(\omega)\coth(\omega/2T)$.

[1] *Semiconductor Spintronics and Quantum Computation*, edited by D. D. Awschalom, D. Loss, and N. Samarth, Series on Nanoscience and Technology (Springer, New York, 2002).
[2] L. N. Bulaevskii and G. Ortiz, Phys. Rev. Lett. **90**, 040401 (2003).
[3] Y. Manassen, R. J. Hamers, J. E. Demuth, and A. J. Castellano, Jr., Phys. Rev. Lett. **62**, 2531 (1989); D. Shachal and Y. Manassen, Phys. Rev. B **46**, 4795 (1992); Y. Manassen, I. Mukhopadhyay, and N. R. Rao, Phys. Rev. B **61**, 16223 (2000).
[4] C. Durkan and M. E. Welland, Appl. Phys. Lett. **80**, 458 (2002).
[5] D. Mozyrsky, L. Fedichkin, S. A. Gurvitz, and G. P. Berman, Phys. Rev. B **66**, 161313(R) (2002).
[6] L. N. Bulaevskii, M. Hruška, and G. Ortiz, Phys. Rev. B **68**, 125415 (2003).
[7] K. A. Matveev, Zh. Eksp. Teor. Fiz. **99**, 1598 (1991) [Sov. Phys. JETP **72**, 892 (1991)]; A. Furusaki and K. A. Matveev, Phys. Rev. B **52**, 16676 (1995).
[8] O. Parcollet and C. Hooley, Phys. Rev. B **66**, 085315 (2002).
[9] J. Paaske, A. Rosch, and P. Wölfle, Phys. Rev. B **69**, 155330 (2004).
[10] A. Kaminski, Yu. V. Nazarov, and L. I. Glazman, Phys. Rev. Lett. **83**, 384 (1999); Phys. Rev. B **62**, 8154 (2000).
[11] A. Rosch, J. Kroha, and P. Wölfle, Phys. Rev. Lett. **87**, 156802 (2001).
[12] J. Paaske, A. Rosch, J. Kroha, and P. Wölfle, Phys. Rev. B **70**, 155301 (2004).
[13] J. R. Ackerhalt and J. H. Eberly, Phys. Rev. D **10**, 3350 (1974).
[14] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions: Basic Processes and Applications* (Wiley, New York, 1992) (Complements Av, pp. 388 ff and C_{IV}, pp. 334 ff).
[15] P. W. Milonni, in *The Quantum Vaccum: An Introduction to Quantum Electrodynamics* (Academic, San Diego, 1994).
[16] C. W. Gardiner and P. Zoller, in *Quantum Noise* (Springer, Berlin, 1999), Chap. 3.
[17] F. Haake, in *Quantum Statistics in Optics and Solid State Physics*, edited by G. Höhler, Springer Tracts in Modern Physics Vol. 66 (Springer, Berlin, 1973), p. 98, Sec. 3b.
[18] G. F. Efremov and A. Yu. Smirnov, Zh. Eksp. Teor. Fiz. **80**, 1071 (1981) [Sov. Phys. JETP **53**, 547 (1981)].
[19] A. Schiller and S. Hershfield, Phys. Rev. B **58**, 14978 (1998).