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Phonon-assisted spin current in a hybrid system with a single molecular quantum dot system applied with ac magnetic fields

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Abstract. We investigate the spin current through a molecular quantum dot (MQD) irradiated with a rotating magnetic field and an oscillating magnetic field by nonequilibrium Green's function. The rotating magnetic field rotates with the angular frequency ω_r around the z-axis with the tilt angle θ, and the time-oscillating magnetic field is located in the z-axis with the angular frequency ω. Different behaviors have been shown in the presence of electron-phonon interaction(EPI) which plays a significant role in the transport. The spin current displays asymmetric behavior as the source-drain bias $eV=0$, novel side peaks or shoulders can be found due to the phonon absorption and emission procedure, and the negative spin current becomes stronger as the parameter g increases. However, the spin currents display the same magnitude and the same oscillation behavior in the region $\mu_0 B_1 > 3\Delta$ regardless the parameter g.

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Key words: molecular quantum dot, spin-flip effect, electron-phonon interaction, nonequilibrium Green's function

1 Introduction

The manipulation of spin is one of the fundamental processes in spintronics, providing the possibility of writing information in a magnetic memory [1], and also because of the possibility of classical or quantum computation using spin. Usually the effect of spin is very small in non-magnetic materials, and it can be neglected in the absence of magnetic field. However, the spin of an electron is responded to an applied magnetic field sensitively. Recently, many efforts have been made to this area. Datta and Das have made

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the pioneering contribution to the exploration of spin-dependent semiconducting nanodevice [2]. In this structure, the current arises from the spin precession due to spin-orbit coupling in narrow-gap semiconductor. The theoretical work of efficient spin filter has been discussed based on a quantum dot (QD) in the Coulomb blockade regime weakly coupled to leads [3]. The spin-current circuit and generator phenomena are also proposed to design spin-battery [4–6]. The Rashba term affects the transport in nonmagnetic resonant tunneling diode [7]. Therefore a very prosperous frontier of investigation known as spintronics is developing correspondingly. The dissipatedness spin transport [8], quantized spin conductance in insulating system provide some examples of spintronics [9]. Spin-flip transport through a quantum dot system also has been discussed [10].

The study of electronic transport through molecular devices has attracted considerable attention [11–14]. Molecular devices consist of single molecules connected to leads, where vibrations and electronic interactions operate [11]. The experiments show that the electron-phonon interaction(EPI) becomes more and more important in electronic transport through a very small single molecular device [11]. As a tunneling electron travels through a MQD, if its residence time on the MQD can be compared with the time of nuclear vibration, the inelastic tunneling will have a great impact on electron transport properties. It was first observed in a single C_{60} molecule that the signs of vibrational sidebands were shown in transport [11, 15–17]. Many other molecules can also be taken as MQDs, such as carbon nanotubes [18], octanethiols [19], ultrasmall metallic particles [20], and other self-assembling organic molecules [21]. Theoretically, many efforts have been made toward quantum transport through MQDs system, such as kinetic equation approach [22], the rate equation approach [23], the nonequilibrium quantum theory [24, 25] and the numerical renormalization group calculation [26, 27]. This paper is contributed to the MQD system under the perturbation of a rotating magnetic field and an oscillating magnetic field, where the spin current is modified due to the EPI. We derive the spin current formula first, and then perform the numerical calculation.

2 Model and formalism

We consider the circumstance that a MQD is coupled to two metallic leads. Only single level MQD without inter- or intradot Coulomb interactions are considered, which is coupled to the local vibration mode and irradiated with a rotating magnetic field and an oscillating magnetic field. The rotating magnetic field rotates with the angular frequency ω_r around the z-axis with the tilt angle θ, and the azimuthal angle $\varphi(t)=\omega_r t$, i. e. , $\mathbf{B}_0(t)=\mathbf{B}_0(\sin\theta\cos\varphi(t))\mathbf{e}_x+\sin\theta\sin\varphi(t))\mathbf{e}_y+\cos\theta\mathbf{e}_z)$. The time-oscillating magnetic field $\mathbf{B}_1(t)$ located in the z-axis is defined as $\mathbf{B}_1(t)=B_1\cos(\omega t)\mathbf{e}_z$, where ω is the angular frequency of the oscillating magnetic field. The total magnetic field applied to the MQD is $\mathbf{B}=\mathbf{B}_0+\mathbf{B}_1$. The Hamiltonian of the system can be written as

$$\begin{aligned} H &= \sum_{\gamma k\sigma}\epsilon_{\gamma,k\sigma}c^\dagger_{\gamma,k\sigma}c_{\gamma,k\sigma}+\sum_{\sigma\sigma'}d^\dagger_\sigma\Omega_{\sigma\sigma'}(t)d_{\sigma'}+\sum_\sigma[\tilde{E}_\sigma(t)+\lambda(a+a^\dagger)]d^\dagger_\sigma d_\sigma \\ &\quad +\sum_{\gamma k\sigma}(V_{\gamma k}c^\dagger_{\gamma,k\sigma}d_\sigma+H.c.)+H_{ph} \end{aligned} \tag{1}$$

where

$$\mathbf{\Omega(t)}=\mathbf{\Omega_0}\begin{pmatrix}\cos\theta & \sin\theta e^{-i\varphi(t)} \\ \sin\theta e^{i\varphi(t)} & -\cos\theta\end{pmatrix} \tag{2}$$

and $\tilde{E}_\sigma(t)=E_\sigma+\lambda_\sigma\mu_0B_1\cos(\omega t)$, where λ_σ is the eigenvalue of the Pauli operaor σ_z, $E_\sigma=E^{(0)}_\sigma+eVg$, $\gamma_0=\mu_0B_0$. The operators $c^\dagger_{\gamma,k\sigma}(c_{\gamma,k\sigma})$, and $d^\dagger_\sigma(d_\sigma)$ are the creation (annihilation) operators of electron in two leads and central MQD, respectively, with $\gamma\in\{L,R\}$. In the Hamiltonian, $\epsilon_{\gamma,k\sigma}$ is the energy of electron in the γth lead, $E^{(0)}_\sigma$ is the energy level of MQD, eV_g is the gate-voltage, λ is the EPI coupling strength , μB_1 is the Zeeman energy and $V_{\gamma k}$ is the coupling strength between the MQD and the γth lead. The last term of Eq. (1) stands for the phonon mode $H_{ph}=\omega_0a^\dagger a$ with the vibrational frequency ω_0 and the creation(annihilation) operator $a^\dagger(a)$.

In order to handle the problem conveniently, we make the gauge transformation $\Psi(t)=\hat{U}(t)\tilde{\Psi}(t)$ over the Schrödinger equation and the Lang-Firsov transformation $\tilde{H}=e^sH'e^{-s}$ over the hamiltonian, where the unitary operator is defined by

$$\begin{aligned} \hat{U}(t) &= exp[-i\Lambda\sum_\sigma\lambda_\sigma sin(\omega t)d^\dagger_\sigma d_\sigma] \\ s &= \lambda/\omega_0\sum_\sigma d^\dagger_\sigma d_\sigma \end{aligned}$$

where $\Lambda=\mu_0B_1/\hbar\omega$. From these two transformation, first, we can remove the time-oscillating Zeeman energy into the interaction terms, second, we can decouple the entanglement of electron and phonon. We get the transformed Hamiltonian

$$\tilde{H}=\sum_{\gamma k\sigma}\epsilon_{\gamma,k\sigma}c^\dagger_{\gamma,k\sigma}c_{\gamma,k\sigma}+\sum_{\sigma\sigma'}d^\dagger_\sigma\tilde{\Omega}_{\sigma\sigma'}(t)d_{\sigma'}+\sum_\sigma\tilde{E}_\sigma d^\dagger_\sigma d_\sigma+\sum_{\gamma k\sigma}(\tilde{V}_{\gamma k}(t)c^\dagger_{\gamma,k\sigma}d_\sigma+H.c.)+H_{ph} \tag{3}$$

In the Hamiltonian Eq. (3), the interaction strengths are changed to the time-dependent ones as $\tilde{V}_{\gamma,k\sigma}(t)=V_{\gamma k}exp[-i\lambda_\sigma\Lambda sin(\omega t)]X$ with $X=exp[-\lambda/\omega_0(a^\dagger-a)]$. The operator X comes from EPI, which can be approximated by its expectation value in the thermal equilibrium, $<X>=exp[-g(N_{ph}+1/2)]$, with $N_{ph}=[e^{\hbar\omega_0/(k_BT)}-1]^{-1}$ [29]. The matrix $\tilde{\mathbf{\Omega}}(t)$ takes the form as in Eq. (1), but with the transformation as $\varphi(t)\rightarrow\tilde{\varphi}(t)=\omega_rt+\alpha_1sin(\omega t)$, where $\alpha_1=2\Lambda$. The energy level of the MQD is renormalized by Lang-Firsov transformation, $\tilde{E}_\sigma=E_\sigma-\Delta$ where $\Delta=g\omega_0$ with $g=(\lambda/\omega_0)^2$.

The tunneling current formula can be derived from the Heisenberg equation and continuity equation by employing the NGF technique. The time averaged current can be expressed as the Landauer-Büttiker-like formula [30]

$$\begin{aligned} I_{\gamma,\sigma\sigma} &= \frac{e}{h}\sum_{knl\beta}\int d\varepsilon L_k T_{\gamma\beta\sigma,nl}(\epsilon-k\hbar\omega_0)\{[f_\gamma(\varepsilon-n\hbar\omega)-f_\beta(\varepsilon-l\hbar\omega)] \\ &\quad +\sum_m J_m^2(\alpha_1)|\tilde{g}^r_{\bar{\sigma}\bar{\sigma}}(\tilde{\varepsilon}_{m\sigma})|^2\gamma(\theta)^2[f_\gamma(\varepsilon-n\hbar\omega)-f_\beta(\tilde{\varepsilon}_{m\sigma}-l\hbar\omega)]\} \end{aligned} \tag{4}$$

where $T_{\gamma\beta\sigma,nl}(\epsilon)=\Gamma_\gamma\Gamma_\beta J_n^2(\Lambda)J_l^2(\Lambda)|\tilde{G}^r_{\sigma\sigma}(\varepsilon)|^2$ represents the transmission coefficient of the electron with spin σ transporting from γth lead to the βth lead. $J_n(x)$ is the Bessel function of the first kind, where $\Lambda=\mu_0 B_1/\hbar\omega$ and $\alpha_1=2\Lambda$.

$$L_k=e^{-g(2N_{ph}+1)}I_k(2g\sqrt{N_{ph}(N_{ph}+1)})e^{n\omega_0\beta/2},$$

$I_k(z)$ is the kth Bessel function of the complex argument. With the help of the equation of motion approach, the diagonal elements of the retarded Green's function for the dressed electron can be evaluated analytically as

$$\tilde{G}^r_{\sigma\sigma}(\epsilon)=\frac{1}{\epsilon-\varepsilon_\sigma(\theta)-\Sigma^r_{\sigma\sigma}(\epsilon)+\gamma(\theta)^2\sum_n J_n^2(\alpha_1)\tilde{g}^r_{\bar{\sigma}\bar{\sigma}}(\tilde{\varepsilon}_{n\sigma})} \tag{5}$$

where $\tilde{\varepsilon}_{n\sigma}=\varepsilon-\lambda_\sigma(\omega_r+n\omega)\hbar$, $\varepsilon_\sigma(\theta)=\tilde{E}_\sigma+\lambda_\sigma\gamma_0\cos\theta$ and $\gamma(\theta)=\gamma_0\sin\theta$. We also have defined the Green's function $\tilde{g}^r_{\sigma\sigma}(\epsilon)=1/[\epsilon-\varepsilon_\sigma(\theta)-\Sigma^r_{\sigma\sigma}(\epsilon)]$. The retarded self-energy is determined by the imaginary part $\Sigma^r_{\sigma\sigma}(\epsilon)=-i\Gamma/2$, where $\Gamma=\Gamma_L+\Gamma_R$. The spin current is defined by

$$I_{\gamma,s}=\hbar(I_{\gamma,\downarrow\downarrow}-I_{\gamma,\uparrow\uparrow}) \tag{6}$$

3 Numerical results

We now discuss numerical results for spin current in molecular quantum dot system with electron-phonon interaction at zero temperature for both the cases as terminal bias $eV=0$ and $eV\neq 0$. Without loss of generality, we consider only the case of a single-level MQD by setting $E_\sigma^{(0)}=0$. Observing a single-level MQD system can tell us about the dominant behavior around the chosen energy level, and the corresponding physical properties can be measured by determining said quantities in the vicinity of this energy level. The energy quantity $\Delta=0.1$ meV is taken as the energy scale for the numerical calculations. We choose the coupling strengths to be symmetrical, with line-width of $\Gamma_L=\Gamma_R=\Gamma=0.005$ meV. The Fermi distribution function becomes the step function at zero temperature as $f_\gamma(\epsilon)=1-\theta(\epsilon-\mu_\gamma)$. The spin curtent is scaled by $I_{s0}=\Delta/(4\pi)$.

The spin current resonance versus the angle θ in the absence of source-drain bias is presented in Fig. 1 to exhibit the modification of spin current by the EPI. The spin

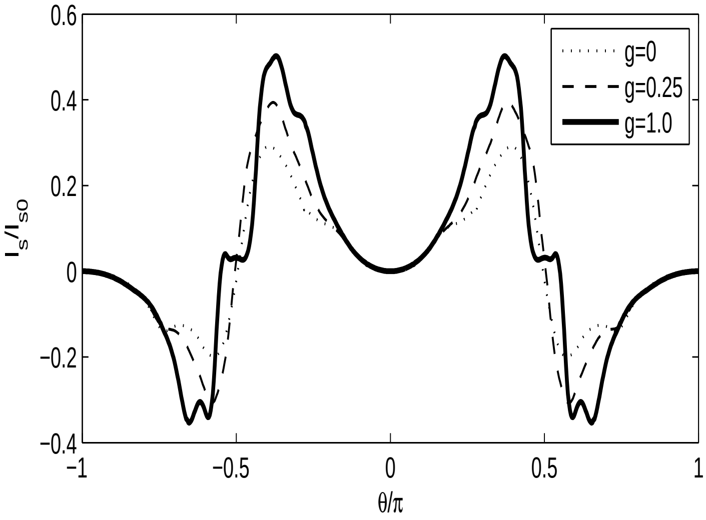

Figure 1: The spin current in unit I_{s0} versus phase θ in the absence of source-drain bias. The parameters are choose as $\hbar\omega_0 = 0.2\Delta$, $\hbar\omega = 0.6\Delta$, $\gamma_0 = \Delta$, $eV = 0$ and $eV_g = 0$. The dotted , dashed, and solid curves correspond to $g = 0,0.25,1.0$, respectively.

current displays symmetric resonant structure. The resonant behavior of the spin current is intimately related to EPI. As the parameter g increases the spin current reflects richer resonance structure, some new shoulders, peaks and valleys emerge. The system provide more channel for transport in the presence of EPI.

We present the spin current with respect to the gate-voltage eV_g in Fig. 2. In the absence of EPI, the spin current caused by the rotating magnetic field and the oscillating magnetic field displays symmetric resonance structure. When the phonon-assisted tunneling turns on, asymmetric resonance structure emerges and becomes more obvious as the parameter g increases. Because at zero temperature no phonon modes are excited on the quantum dot, the electrons tunneling onto the dot can excite phonon modes, and phonon-emission processes are allowed. At zero temperature, L_n, which relates with EPI, simply reads

$$L_n \equiv \begin{cases} e^{-g} g^n / n! & n \geq 0 \\ 0 & n < 0 \end{cases} .$$

Negative spin current emerges around $eV_g = \Delta$ in the presence of EPI, and it becomes bigger as the parameter g increases.

Fig. 3 displays the spin current in the presence of EPI varying with the magnitude of the oscillating magnetic field $\mu_0 B_1$. The spin current is strongly dependent on the magnitude of the oscillating magnetic field and the coupling strength of EPI. The spin current varies nonmonotonically in the whole region $\mu_0 B_1$. There exists a peak in the spin current by changing the B_1 for each parameter g. The magnitude of the peak is strongly

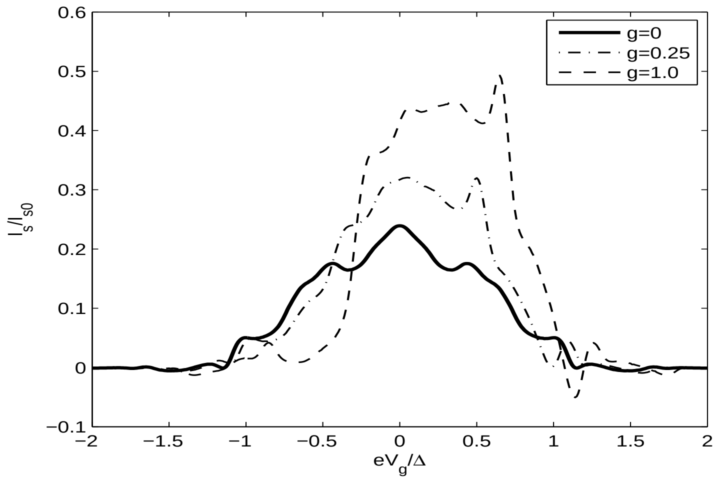

Figure 2: The spin current in unit I_{s0} versus gate voltage eV_g in the absence of source-drain bias. The parameters are choose as $\hbar\omega_0=0.2\Delta$, $\hbar\omega=0.6\Delta$, $\gamma_0=\Delta$ and $eV=0$. The dotted , dashed, and solid curves correspond to $g=0,0.25,1.0$, respectively.

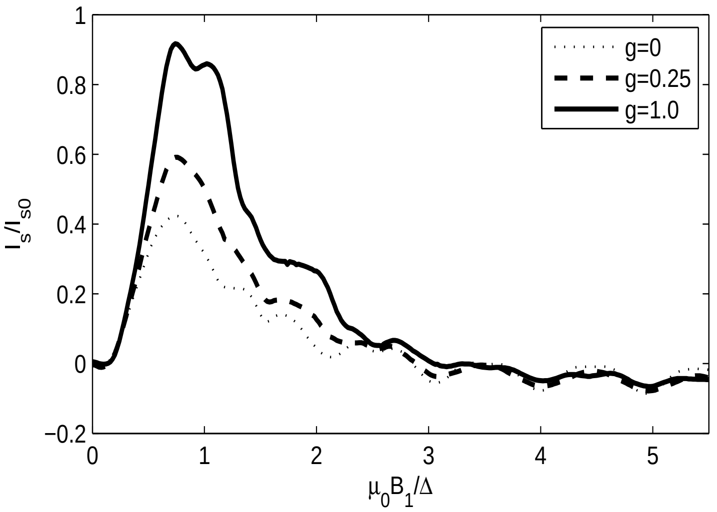

Figure 3: The spin current in unit I_{s0} versus phase θ in the absence of source-drain bias. The parameters are choose as $\hbar\omega_0=0.2\Delta$, $\hbar\omega=0.6\Delta$, $\gamma_0=\Delta$, $eV=0.5$ and $eV_g=0$. The dotted , dashed, and solid curves correspond to $g=0,0.25,1.0$, respectively.

determined by the parameter g, however the location of the peak is about 0.8Δ for each parameter g. The magnitude of the peak increases as the parameter g increases. However, the spin currents display the same magnitude and the same oscillation behavior in the region $\mu_0 B_1 > 3\Delta$.

4 Summary

We have investigated the spin current for the system of vibrating quantum dot irradiated with a rotating magnetic field and an oscillating magnetic field. The current is derived from the equation of motion method incorporated with the nonequilibrium Green's function of QD. The spin current is driven by the rotating magnetic field and the oscillating magnetic field. The information of EPI is transferred to the spin current. The spin current displays richer resonance structure in the presence of EPI. The symmetric spin current is destroyed by the EPI. Also the system reflects negative spin current due to the EPI. However, as the magnitude of oscillating magnetic field $\mu_0 B_1 > 3\Delta$ the spin current displays the same behavior regardless the parameter g.

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