Spin current and shot noise in single-molecule quantum dots with a phonon mode

Hui Yu and J.-Q. Liang
Institute of Theoretical Physics, Shanxi University, Taiyuan, Shanxi 030006, China
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In this paper, we investigate the spin-current and its shot-noise spectrum in a single-molecule quantum dot coupled with a local phonon mode. We pay special attention to the effect of the phonon on the quantum transport property. The spin-polarization-dependent current is generated by a rotating magnetic field applied in the quantum dot. Our results show the remarkable influence of phonon mode on the zero-frequency shot noise. The electron-phonon interaction leads to sideband peaks that are located exactly on the integer number of the phonon frequency, and moreover, the peak height is sensitive to the electron-phonon coupling.

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I. INTRODUCTION

Modern nanotechnology has provided the possibility to fabricate electronic devices in which the acting element is a single, organic molecule. This has led to a growing interest in the study of transport properties of molecular devices.[1–3] Such a device may be modeled as a quantum dot (QD) weakly coupled to the macroscopic charge reservoirs. In addition to their practical applications, these artificial, tunable devices, such as electronic components,[1] Coulomb blockade structures,[4] diodes,[5] or switching devices with high negative differential resistance,[6] are important for understanding the basic physics, including the many-body and the size effects. In contrast to the semiconductor QDs, molecules (the linear size is at least one order smaller than that of the former) are intrinsically different from semiconductor nanostructures. The devices with molecules may lead to different physics, especially when electrons are added or removed from a single molecule. Since the molecular material possesses much smaller elastic parameters, it is very easy to excite the internal, vibrational degrees of freedom[6–11] (phonon modes) when electrons are incident upon the molecules through a tunnel junction. Thus, molecules react inevitably to the tunnel electrons even at low temperature. This phenomenon has provoked a large amount of experimental[3,8,10–15] investigations into the problem of transport through mesoscopic systems with electron-phonon coupling. Inelastic scattering effects have been observed directly in measurements of the differential conductance of molecules adsorbed on metallic substrates with scanning tunneling microscopy[3] (STM). In a series of experiments by Park *et al.*[8] it was shown that the current through a single C_{60} molecule was strongly influenced by the vibrational mode. Zhitenev *et al.*[11] have also demonstrated that the low-bias conductance of molecules is dominated by resonant tunneling through coupled electronic and vibrational levels. There have been a number of theoretical efforts[14–25] focused on the effects of electron-phonon (E-PH) coupling in mesoscopic systems, with basic models capturing the essential physics and standard methods. Various aspects of the electron-phonon/vibron interaction effect on the tunneling through molecule QDs have been studied by many authors.

On the other hand, motivated by the easy control of electron spin as well as the remarkably long coherence time, the spin-polarization-dependent transports in open QDs[26–28] have attracted considerable attention. These spin-source devices not only exhibit the fundamental physics but also promise applications in the technologies of spintronics and quantum information.[29] A pure spin current has been reported by direct optical injection without generating a net charge current.[30] Theoretically, there are number of mechanisms proposed to produce pure spin current[31–33] using techniques of ferromagnetic resonance in a ferromagnetic-normal-metal[31] or electron-spin resonance (ESR) in a QD-based system with sizable Zeeman splitting.[32]

Moreover, a trend in the transport studies in mesocopic systems is toward not only considering the transport characteristics of a given device but also examining the noise properties. Due to the discrete nature of charge carriers, electrical current through a conductor is subject to time-dependent fluctuation around its mean value, which manifests the consequence of the quantization of the charge carriers and is usually referred to as the shot noise in literature.[35] Shot noise defined as the mean-square fluctuations of the current flowing through a given terminal at zero temperature is of great importance and interest, because the spectrum of shot noise contains additional information about the interactions that the conduction electrons undergo[34] beyond the mean-current properties and can be used to discern different mechanisms resulting in the same mean current. For example, shot-noise experiments can determine the kinetics of electrons and reveal information about the correlation of electronic wave functions. Thus, shot noise has been extensively studied in a wide variety of systems.[34–39]

Furthermore, the spin-resolving current correlation is more useful to describe electron correlation, because the electronic wave packet with opposite spins is uninfluenced by the Pauli exclusion principle and only reflects unambiguous information about the interaction. For a two-lead device, correlations can be formulated by quantities measured at the same lead, i.e., the autocorrelation, or by quantities measured at the two different leads, namely, the cross-correlation. Büttiker[36] pointed out that while cross-correlations of the charge noise can either be positive or negative for bosons, they are necessarily negative definite for fermions in both the equilibrium and the transport regimes. It is well known[36] that antibunching in a fermionic system gives rise to negative definite cross-correlation for the charge current. The conser-

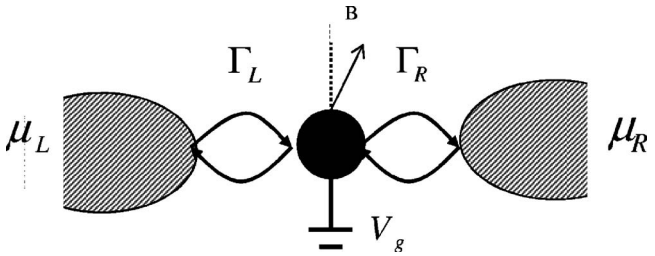

FIG. 1. Schematic diagram of a molecule quantum-dot system.

vation of charge forces the cross-correlation and autocorrelation of the charge-current noise spectra to differ by just a minus sign in a two-probe system. However, for a pure spin current, the situation is very different because of a lack of spin-current conservation due to the spin flipping. The autocorrelation of the spin-current is surely positive definite, while the cross-correlation is either positive or negative, depending on a number of parameters.[39] The zero-frequency shot noise $S(0)$ for a classical conductor[40] is characterized by the Poisson value $S_p(0)=2e\langle I\rangle$, where $\langle I\rangle$ is the average current, while the shot noise in a noninteracting mesoscopic conductor is always reduced by the Pauli exclusion in comparison with the Poisson value. Of course, shot noise is also influenced by other factors, such as electron-electron and electron-phonon interactions.

Motivated by the achievements in the single-molecule and spin-current experiments, in this paper, we will, based on the ESR mechanism[39,41] of generating net spin current,[32] investigate theoretically the electron-phonon effects on the spin current and its shot noise for molecular dots. We use the Keldysh nonequilibrium Green's-function technique to calculate the spin current and shot noise through a single-molecule coupled to electron reservoirs and focus on the effect of the inelastic scattering process. The ESR-type model with a single dispersionless phonon mode is employed to address the vibrational degrees of freedom in the molecular dot. All other complexity of real molecular devices, apart from interaction with a single longitudinal-optical (LO) phonon localized on the molecule, is ignored.

II. MODEL AND HAMILTONIAN

The model system under consideration is illustrated in Fig. 1, which consists of a molecule of one relevant level coupled with a single (Einstein) phonon mode, and two leads, which we label as "left" and "right." A time-dependent (rotating) magnetic field $\mathbf{B}(t)$ is applied in the molecule dot to flip the spin of the electron. Also, a gate electrode is capacitively attached to the dot to tune the energy level. The total Hamiltonian of the system is written as

$$H = H_L + H_R + H_P + H_D + H'(t) + H_T, \tag{1}$$

where

$$H_L + H_R = \sum_{k\sigma,\alpha=L,R} \varepsilon_k C^+_{k\alpha\sigma} C_{k\alpha\sigma}, \tag{2}$$

$$H_P = w_0 a^+ a, \tag{3}$$

$$H_D = \sum_{\sigma} [(\varepsilon + \sigma B_0 \cos\theta) + \lambda(a + a^+)] d^+_\sigma d_\sigma, \tag{4}$$

$$H'(t) = r[\exp(-iwt) d^+_\uparrow d_\downarrow + \exp(iwt) d^+_\downarrow d_\uparrow], \tag{5}$$

with $r=B_0 \sin\theta$

$$H_T = \sum_{k,\sigma,\alpha=L,R} (T_{k\alpha} C^+_{k\alpha\sigma} d_\sigma + \text{c.c.}). \tag{6}$$

The first two terms H_L and H_R of Eq. (1) are, respectively, the Hamiltonians for electrons in the left and right noninteracting metallic leads, where $C^+_{k\alpha\sigma}(C_{k\alpha\sigma})$ are the creation (annihilation) operators of electrons with momentum k, spin σ, and energy ε_k in the lead α. Here, we have set the same chemical potential for both leads. The third term H_P describes the nondispersive, LO phonon, where w_0 is the frequency of the single-phonon mode, and $a^+(a)$ is the phonon creation (annihilation) operator. H_D and $H'(t)$ correspond to the interaction Hamiltonians between the electron and phonon in the QD, which is subjected to a time-dependent magnetic field with uniform strength,

$$\mathbf{B}(t) = B_0(\sin\theta\cos\omega t,\ \sin\theta\sin\omega t,\ \cos\theta),$$

where B_0 is the constant field strength. Here, $d^+_\sigma(d_\sigma)$ are the electron creation (annihilation) operators in the QD, and $\varepsilon = \varepsilon_0 + eV_g$ is the single energy level of the molecule, which can be controlled by the gate voltage V_g, where e denotes the absolute value of the electron charge. λ is the coupling constant between the electron in the molecule dot and the LO phonon mode with energy w_0. H_T represents the coupling of the molecule with leads, where the tunneling matrix elements $T_{k\alpha}$ transfer electrons through an insulating barrier out of the dot.

A. Spin-current and shot-noise formula

We define the spin-dependent particle-current operator in the lead α as ($\hbar=1$)

$$\hat{J}_{\alpha,\sigma\sigma'} \equiv \sum_k \frac{d(C^+_{k\alpha\sigma} C_{k\alpha\sigma'})}{dt} = -i\sum_k (T_{k\alpha} C^+_{k\alpha\sigma} d_{\sigma'} - T^*_{k\alpha} d^+_\sigma C_{k\alpha\sigma'}). \tag{7}$$

The spin-current operator of the spin component σ is then

$$J_{s\alpha} = \frac{1}{2}\sum_{\sigma\sigma'} J_{\alpha,\sigma\sigma'} \boldsymbol{\sigma}^z_{\sigma\sigma'}, \tag{8}$$

where $\boldsymbol{\sigma}^z$ is the Pauli matrix.

The spin-dependent current can be computed from the current operator [Eq. (7)]

$$I_{\alpha,\sigma\sigma'}(t) \equiv \langle \hat{J}_{\alpha,\sigma\sigma'}(t)\rangle = -\sum_k [T_{k\alpha} G^<_{d\sigma,k\alpha\sigma'}(t,t) - T^*_{k\alpha} G^<_{k\alpha\sigma',d\sigma}(t,t)], \tag{9}$$

where the nonequilibrium Green's functions (NEGFs) are defined as

$$G^{<}_{d\sigma,k\alpha\sigma'}(t,t') \equiv i\langle C^{+}_{k\alpha\sigma'}(t')d_{\sigma}(t)\rangle,$$

$$G^{<}_{k\alpha\sigma,d\sigma'}(t,t') \equiv i\langle d^{+}_{\sigma'}(t')C_{k\alpha\sigma}(t)\rangle.$$

Using the Keldysh nonequilibrium Green's function formalism,[37] we obtain the spin-dependent current

$$I_{\alpha,\sigma\sigma'} = \frac{1}{2N_\tau}\int \frac{dE_1}{2\pi}\int \frac{dE_2}{2\pi}\Gamma_\alpha\Gamma\sum_{\sigma_2} G^{r}_{\sigma\sigma_2}(E_1,E_2)G^{a}_{\sigma_2\sigma'}(E_2,E_1) \times[f(E_2)-f(E_1)], \tag{10}$$

where

$$\Gamma = \sum_{\alpha=L,R}\Gamma_\alpha$$

is the total tunnel-coupling constant, which is a function of energy E, and $G^{r(a)}_{\sigma\sigma'}(E_1,E_2)$ are the Fourier transforms of the dot-electron retarded (advanced) Green's function

$$G^{r(a)}_{\sigma\sigma'}(t,t') = \mp i\theta(\pm t \mp t')\langle[d_\sigma(t),d^{+}_{\sigma'}(t')]\rangle$$

in the presence of both the electron-phonon interaction and the tunneling coupling between dot and leads, $\mathbf{\Gamma}_\alpha(E)$, the elastic coupling to the α lead (referred to as the linewidth function) depends on the hopping strength and the density of states $\rho_\alpha(E)$ in the lead α according to

$$\Gamma_\alpha(E) \equiv 2\pi\rho_\alpha(E)|T_{k\alpha}(E)|^2. \tag{11}$$

In the wide-band limit[42] in which the bandwidth in the leads is much larger than both the resonance width and phonon energies, the contact densities of states are constant in the region of the resonance. If the hopping matrix elements also vary slowly with energy, the couplings with the contacts Γ_α are independent of energy as well. The Fermi distribution of the lead α is

$$f_\alpha(E) = \{\exp[(E-\mu_\alpha)/kT]+1\}^{-1}. \tag{12}$$

The noise spectra of both the charge current and the spin current can be obtained from the correlation $S^{\sigma\sigma'}_{\alpha\beta}$ between the spin-dependent particle currents in leads α and β

$$S^{\sigma\sigma'}_{\alpha\beta} = \langle[\hat{J}_{\alpha\sigma}(t_1)-\langle\hat{J}_{\alpha\sigma}(t_1)\rangle][\hat{J}_{\beta\sigma'}(t_2)-\langle\hat{J}_{\beta\sigma'}(t_2)\rangle]\rangle. \tag{13}$$

Here $\langle\cdots\rangle$ denotes both the statistical average and the quantum average on the nonequilibrium state.

Briefly, we substitute Eq. (7) and Eq. (9) into Eq. (13) and apply the analytic continuation[43] so that the exact expression of the zero-frequency, spin-dependent correlation can be obtained. It has been shown in Ref. 39 that both the cross-correlation and the autocorrelation are needed to characterize the shot noise of the spin current for the two-lead system, because a spin current is not conserved due to the spin flip induced by the rotating magnetic field. The cross-correlation shot noise of the spin current is

$$S_{LR} \equiv S_{spin,1} = \langle(\Delta J_{L\uparrow}-\Delta J_{L\downarrow})(\Delta J_{R\uparrow}-\Delta J_{R\downarrow})\rangle = \int \frac{dE}{8\pi}f_\downarrow(1-f_\uparrow)\Gamma_L\Gamma_R[|G^{r}_{\uparrow\downarrow}|^2+|G^{r}_{\downarrow\uparrow}|^2 - 2\Gamma^2(|G^{r}_{\uparrow\downarrow}|^4+|G^{r}_{\downarrow\uparrow}|^4)], \tag{14}$$

and the autocorrelation shot noise is defined by

$$S_{LL} \equiv S_{spin,2} = \langle(\Delta J_{L\uparrow}-\Delta J_{L\downarrow})(\Delta J_{L\uparrow}-\Delta J_{L\downarrow})\rangle = \int \frac{dE}{8\pi}f_\downarrow(1-f_\uparrow)\{2[-\Gamma_L^2\Gamma^2(|G^{r}_{\uparrow\downarrow}|^4+|G^{r}_{\downarrow\uparrow}|^4) + \Gamma_L\Gamma(|G^{r}_{\uparrow\downarrow}|^2+|G^{r}_{\downarrow\uparrow}|^2)]-\Gamma_L\Gamma_R(|G^{r}_{\uparrow\downarrow}|^2+|G^{r}_{\downarrow\uparrow}|^2)\}, \tag{15}$$

where $f_\uparrow = f_\uparrow(E)$ and $f_\downarrow = f_\downarrow(E-w)$.

Once the retarded Green's functions $G^{r(a)}_{\sigma\sigma'}(E_1,E_2)$ are known, the spin current and shot noise can be calculated using Eqs. (10) and (13)–(15). In the following, we calculate $G^{r(a)}_{\sigma\sigma'}(E_1,E_2)$ with the standard Dyson equation approach. To this end, we regard the term that explicitly depends on time **t** in the Hamiltonian (1) as the interacting part H_I, such that $H_0 \equiv H - H_I$. Denoting Green's functions for the Hamiltonian H_0 as $G^{0r}_{\sigma\sigma'}(\varepsilon)$, the full Green's functions for Hamiltonian (1) are then calculated from the Dyson equation

$$\mathbf{G}^{r}(E_1,E_2) = 2\pi\mathbf{G}^{0r}(E_1)\delta(E_1-E_2) + \int \frac{dE}{2\pi}\mathbf{G}^{r}(E_1,E+E_2)\mathbf{H}'(E)\mathbf{G}^{0r}(E_2), \tag{16}$$

where the boldface notation indicates that the electron Green's function in the QD and the interacting Hamiltonian $\mathbf{H}'$ are the 2×2 matrices in the spin space, where the element $H'_{\sigma_1\sigma_2}(E)$ is the Fourier transform of $H'_{\sigma_1\sigma_2}(t)$, which is seen to be

$$H'_{\sigma_1\sigma_2}(t) = r(e^{-iwt}d^{+}_{\uparrow}d_{\downarrow} + e^{iwt}d^{+}_{\downarrow}d_{\uparrow}). \tag{17}$$

The full retarded Green's functions of Hamiltonian (1) are then obtained from Eq. (16), after tedious but straightforward algebra, explicitly as (Ref. 24)

$$G^{r}_{\sigma\sigma}(E_1,E_2) = \frac{2\pi\delta(E_1-E_2)G^{0r}_{\sigma\sigma}(E_1)}{1-r^2g(E_1)}, \tag{18}$$

$$G^{r}_{\sigma\bar{\sigma}}(E_1,E_2) = 2\pi\delta(E_1+\bar{\sigma}w-E_2)\frac{rg(E_1)}{1-r^2g(E_1)}, \tag{19}$$

where $g(E_1) \equiv G^{0r}_{\sigma\sigma}(E_1)G^{0r}_{\sigma\bar{\sigma}}(E_1+\bar{\sigma}w)$, $\bar{\sigma}=-\sigma$. Using these relations, it is straightforward to derive the expression of the spin current and shot noise from Eq. (10) and Eqs. (13)–(15).

Since we are interested in the case of strong strength of E-PH interaction, the Green's function $G^{0r}_{\sigma\sigma'}(E)$ can be calculated by performing a standard canonical transformation, $\tilde{H} = e^{s}He^{-s}$, with $S = (\lambda/w_0)\Sigma_\sigma d^{+}_\sigma d_\sigma(a^{+}-a)$.[44] Then all transformed operators are seen to be

$$\tilde{d}_\sigma = d_\sigma X,$$

$$\tilde{d}_\sigma^+ = d_\sigma^+ X^+, \tag{20}$$

$$\tilde{a} = a - \frac{\lambda}{w_0}\sum_\sigma d_\sigma^+ d_\sigma,$$

$$\tilde{a}^+ = a^+ - \frac{\lambda}{w_0}\sum_\sigma d_\sigma^+ d_\sigma, \tag{21}$$

with

$$X = \exp\left[-\frac{\lambda}{w_0}(a^+ - a)\right]. \tag{22}$$

The electron number operator in the QD is invariant under the transformation,

$$\tilde{d}_\sigma^+ \tilde{d}_\sigma = d_\sigma^+ d_\sigma. \tag{23}$$

Then the transformed Hamiltonian can be written as

$$\tilde{H} = \tilde{H}_L + \tilde{H}_R + \tilde{H}_X + \tilde{H}_D + \tilde{H}'(t) + \tilde{H}_T, \tag{24}$$

where

$$\tilde{H}_L + \tilde{H}_R = \sum_{k\sigma,\alpha=L,R} \varepsilon_k C_{k\alpha\sigma}^+ C_{k\alpha\sigma},$$

$$\tilde{H}_X = w_0\left(a^+ - \frac{\lambda}{w_0}\sum_\sigma d_\sigma^+ d_\sigma\right)\left(a - \frac{\lambda}{w_0}\sum_\sigma d_\sigma^+ d_\sigma\right),$$

$$\tilde{H}_D = \sum_\sigma \left[(\varepsilon + \sigma B_0 \cos\theta) + \lambda\left(a + a^+ - 2\frac{\lambda}{w_0}\sum_{\sigma'} d_{\sigma'}^+ d_{\sigma'}\right)\right] d_\sigma^+ d_\sigma,$$

$$\tilde{H}'(t) = r[\exp(-iwt) d_\uparrow^+ d_\downarrow + \exp(iwt) d_\downarrow^+ d_\uparrow],$$

$$\tilde{H}_T = \sum_{k,\sigma,\alpha=L,R} (T_{k\alpha} C_{k\alpha\sigma}^+ d_\sigma X + c.c.). \tag{25}$$

The hopping terms between the molecule and leads [Eq. (25)] are modified by a factor X, which describes the fact that the electron hopping is accompanied by a phonon cloud. Here, to avoid unnecessary complication, we consider the leads that are unaffected by the phonons. This means that we ignore a factor that results from the average of the X operator and does not lead to qualitative changes of the tunneling current. The justification for this is given in Refs. 16 and 44. Consequently, we have

$$\tilde{H}_T \approx H_T,$$

and

$$\tilde{H} = \tilde{H}_{el} + \tilde{H}_{ph}, \tag{26}$$

where

$$\tilde{H}_{el} = \sum_{k\sigma,\alpha=L,R} \varepsilon_k C_{k\alpha\sigma}^+ C_{k\alpha\sigma} + \sum_\sigma (\varepsilon + \sigma B_0 \cos\theta - \Delta) d_\sigma^+ d_\sigma + \sum_{k,\sigma,\alpha=L,R} (T_{k\alpha} C_{k\alpha\sigma}^+ d_\sigma + c.c.) + r[\exp(-iwt) d_\uparrow^+ d_\downarrow + \exp(iwt) d_\downarrow^+ d_\uparrow],$$

$$\tilde{H}_{ph} = w_0 a^+ a,$$

with $\Delta = \lambda^2 / w_0$. Due to the E-PH interaction, the single-energy level of the molecule is renormalized to $\varepsilon' = \varepsilon - \Delta$, and then the Green's function $G_{\sigma\sigma'}^{0r}(t)$ can be decoupled as

$$G_{\sigma\sigma'}^{0r}(t) = -i\theta(t)(\langle \tilde{d}_\sigma(t) \tilde{d}_{\sigma'}^+(0)\rangle_{el} \langle X(t) X^+(0)\rangle_{ph} - \langle \tilde{d}_{\sigma'}^+(0) \tilde{d}_\sigma(t)\rangle_{el} \langle X^+(0) X(t)\rangle_{ph}), \tag{27}$$

where

$$\tilde{d}_\sigma(t) = e^{i\tilde{H}_{el}t} d_\sigma e^{-i\tilde{H}_{el}t},$$

$$X(t) = e^{i\tilde{H}_{ph}t} X e^{-i\tilde{H}_{ph}t}.$$

The renormalization factor due to the E-PH interaction is evaluated as (Ref. 44)

$$\langle X(t) X^+(0)\rangle_{ph} = e^{-\Phi(t)},$$

$$\langle X^+(0) X(t)\rangle_{ph} = e^{-\Phi(-t)},$$

where

$$\Phi(t) = g[N_{ph}(1 - e^{iw_0 t}) + (N_{ph} + 1)(1 - e^{-iw_0 t})], \tag{28}$$

with parameters $N_{ph} = 1/[\exp(\beta w_0) - 1]$ and $g = (\lambda / w_0)^2$.

The Fourier transform of the Green's function $G_{\sigma\sigma'}^{0r}(E)$ is given by

$$G_{\sigma\sigma}^{0r}(E) = \exp[-g(2N_{ph} + 1)] \times \sum_{l=-\infty}^{\infty} I_l[2g\sqrt{N_{ph}(N_{ph} + 1)}] \exp^{(lw_0/2)\beta} \times \exp^{-iw_0 lt} \cdot [(1 - \langle n_{d,\sigma}\rangle) \tilde{G}_{\sigma\sigma}^{0r}(E - lw_0) + \langle n_{d,\sigma}\rangle \tilde{G}_{\sigma\sigma}^{0r}(E + lw_0)], \tag{29}$$

where $\tilde{G}_{\sigma\sigma}^{0r}(E)$ is the retarded Green's function corresponding to the time-independent part of the new Hamiltonian $\tilde{H}_{el}$, the index l indicates the number of phonons involved, and $\langle n_{d,\sigma}\rangle$ is the time-averaged electron-occupation number in the molecule. Here, we consider the case of an "empty" QD, i.e., $\langle n_{d,\sigma}\rangle = 0$. With a little algebra, we find

$$\tilde{G}_{\sigma\sigma'}^{0r}(E) = \frac{\delta_{\sigma\sigma'}}{E - (\varepsilon + \sigma B_0 \cos\theta - \Delta) - \Sigma_{\sigma\sigma'}^r}, \tag{30}$$

where the retarded self-energy due to the tunneling into the electrical leads is given by

$$\Sigma^r_{\sigma\sigma'}(E) = \sum_{k,\alpha\in L,R} \frac{|T_{k\alpha}|^2 \delta_{\sigma\sigma'}}{E - \varepsilon_k + i0^+} = \Lambda(E) - \frac{i}{2}\Gamma(E). \quad (31)$$

In the wide-band limit, the level shift $\Lambda(E)$ can be neglected, and the linewidths are energy-independent constants. Thus, the retarded self-energy can be expressed as

$$\Sigma^r_{\sigma\sigma'}(E) = -\frac{i}{2}\Gamma. \quad (32)$$

The Fourier transform of the full Green's function given by Eq. (27) can be obtained as

$$G^{0r}_{\sigma\sigma'}(E) = \exp[-g(2N_{ph}+1)] \cdot \sum_{l=-\infty}^{\infty} I_l[2g\sqrt{N_{ph}(N_{ph}+1)}] \times \frac{e^{w_0 l\beta/2}}{E - (\varepsilon + \sigma B_0 \cos\theta - \Delta) - lw_0 + \frac{i}{2}\Gamma} \quad (33)$$

III. NUMERICAL RESULTS AND DISCUSSION

We now present the numerical results of the spin current and zero-frequency shot noise. For simplicity, we consider the symmetric tunnel coupling between the molecule and the two leads, i.e., $\Gamma_L=\Gamma_R=\Gamma/2$, and further assume that the energy level of the molecule is controlled by the gate voltage V_g such that $\varepsilon(V_g)=\varepsilon_0+eV_g$, where ε_0 denotes the single-electron energy in the molecule in the absence of the gate voltage. The phonon energy is chosen as the energy unit throughout the rest of this paper. We also set $\hbar=e=1$. In Fig. 2, we plot the spin current I_s vs the parameter r [Fig. 2(a)], the gate voltage V_g [Fig. 2(b)], and the frequency w of the rotating magnetic field [Fig. 2(c)] respectively, at zero temperature $T=0$. The parameter values used in Fig. 2(a) are such that $\Gamma=0.04$, $V_g=0$, $g=0.5^2$, and $\theta=88°$. For comparison, we also plot the spin current in the absence of the E-PH interaction (dashed line). It is clearly shown in Fig. 2(a) that the E-PH interaction results in the shift of resonance peaks. Figure 2(b) shows the spin current I_s as a function of the gate voltage V_g for different r, with $\Gamma=0.04$, $g=0.6^2$, $w=0.1$, and

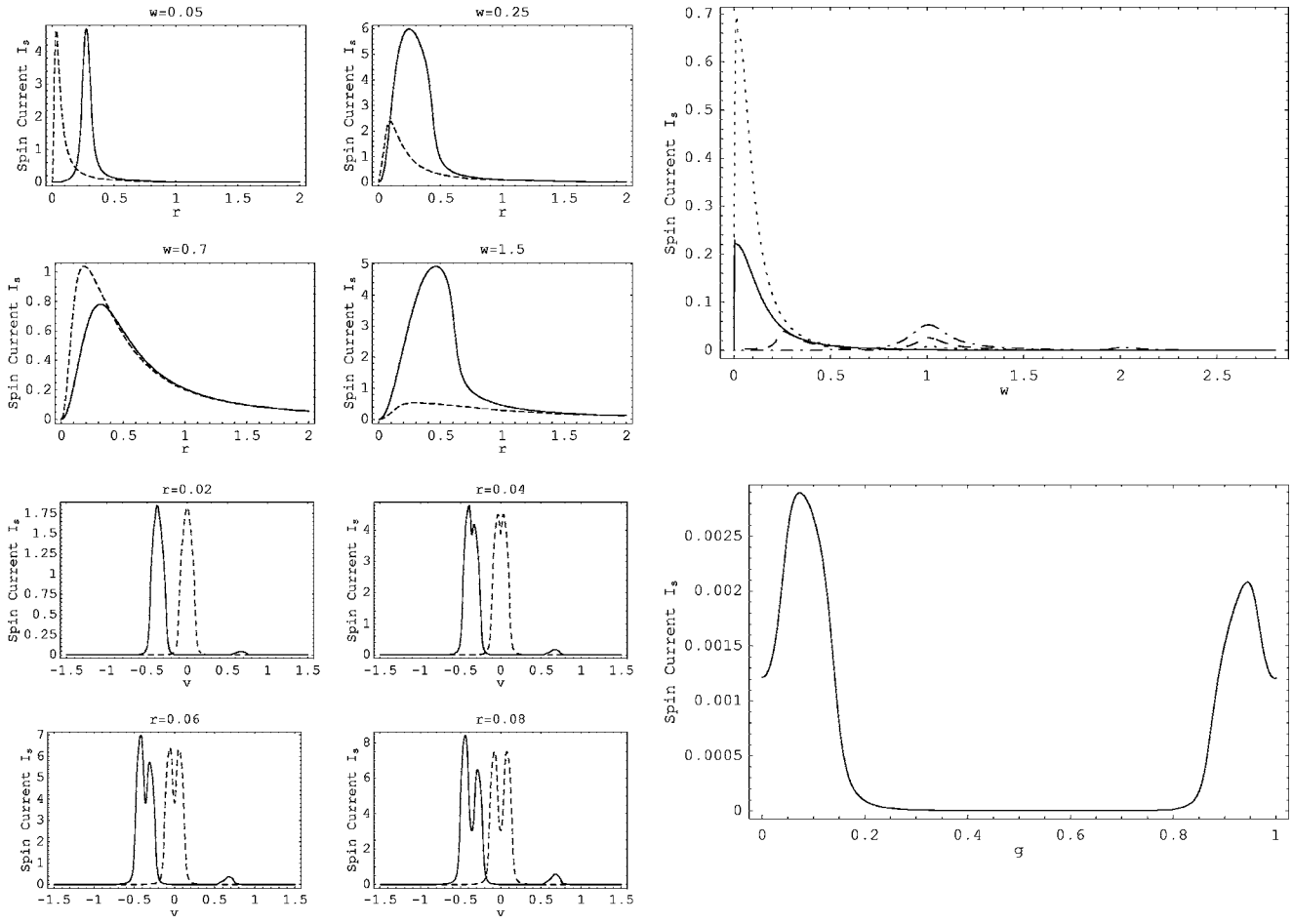

FIG. 2. (a) The spin current I_s vs the parameter r ($\Gamma=0.04$, $V_g=0$, $g=0.5^2$, and $\theta=88°$, with frequencies of the magnetic field $w=0.05$, 0.25, 0.7, and 1.5) for the two cases with (solid line) and without (dashed line) the phonon mode for comparison. (b) The spin current I_s as a function of the gate voltage V_g ($\Gamma=0.04$, $g=0.6^2$, $w=0.1$, and $\theta=88°$, with the parameters $r=0.02$, 0.04, 0.06, and 0.08) for the two cases with (solid line) and without (dashed line) the phonon mode for comparison. (c) The spin current I_s/w vs the frequency w of the rotating magnetic field, with $\Gamma=0.04$, $V_g=0$, $r=0.08$, and $\theta=88°$ for various coupling constants $g=0$ (solid curve), $g=0.3^2$ (dotted curve), $g=0.5^2$ (dashed curve), and $g=0.7^2$ (dashed-dotted curve). (d) The spin current I_s as a function of the coupling constant g.

$\theta=88°$. In the presence of E-PH coupling, the overall spectrum is shifted by a quantity $\Delta=\lambda^2/w_0$ toward the negative gate voltage region. In addition to the main peak, which is related to the molecule energy level, small satellite resonant peaks appear at the positive-energy side. At zero temperature, only phonon-emission processes are allowed, since before tunneling, the phonon state is a vacuum, which explains why the satellite peaks are located at the positive-energy region. Moreover, the height of the satellite peaks is much smaller than the main resonant peak because of the suppression by the E-PH coupling. Figure 2(c) displays the spin current I_s vs the frequency w of the rotating magnetic field for various coupling constants $g=0,0.3^2,0.5^2,0.7^2$ (corresponding to the solid, dotted, dashed, and dashed-dotted curves), with $\Gamma=0.04$, $V_g=0$, $r=0.08$, and $\theta=88°$. In the absence of the phonon mode, only one resonant peak exists. The E-PH coupling typically leads to the new satellite resonant peaks. It is also seen that the heights of the satellite peaks increase with the coupling constant g. The positions of the side peaks are located in $w=nw_0$ $(n=1,2,3,4,\ldots)$. Finally, the plot of the spin current I_s as a function of the E-PH coupling constant g [Fig. 2(d)] shows a double maximum.

Figure 3 shows the dependence of cross-correlation shot noise on the parameter r [Fig. 3(a)], the gate voltage V_g [Fig. 3(b)], and the frequency w of the rotating magnetic field [Fig. 3(c)], respectively, with the same parameter values as in Fig. 2. We see in Fig. 3 that the cross-correlation shot noise of the spin current displays very different behavior from the spin current itself; therefore, the measurements of the shot-noise spectrum can provide more information about the transport properties in mesoscopic systems. In Fig. 3(a), it can be observed that in the absence of the E-PH interaction, the cross-correlation shot noise can be either positive or negative, as r is changed due to the competition[27] between the cross-correlations of electrons with parallel and antiparallel spins. The E-PH interaction has considerable influence on the cross-correlation shot noise such that the negative shot noise is substantially suppressed and even vanishes. Figure 3(b) shows that with r increasing, the cross-correlation shot-

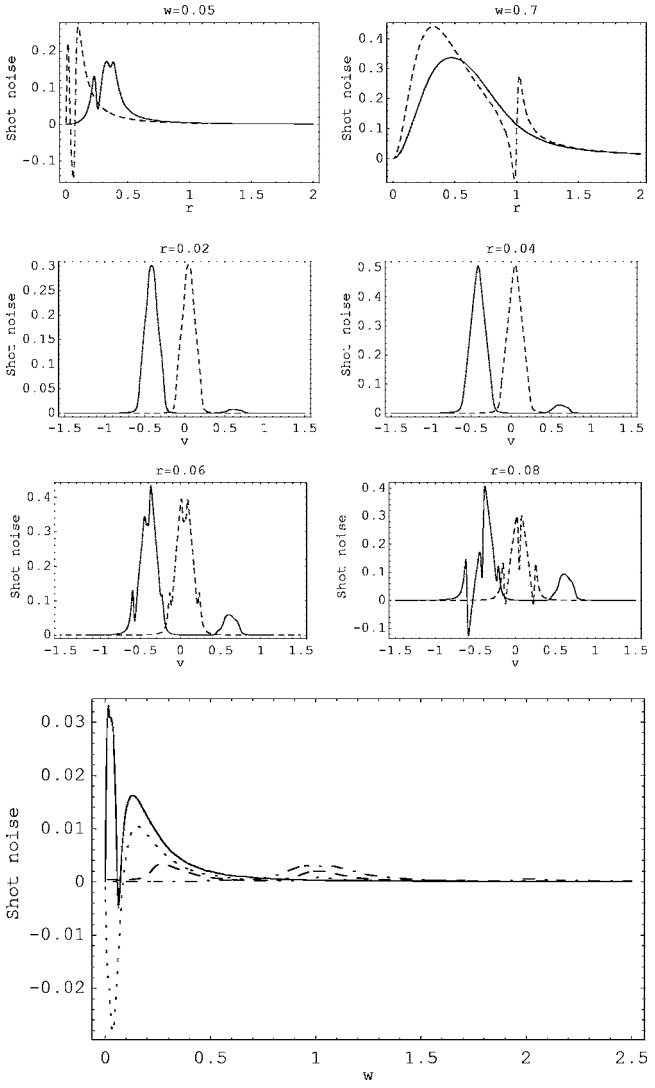

FIG. 3. (a) The cross-correlation shot noise as a function of the parameter r (solid line, with phonon mode, dashed line, without phonon mode). (b) The cross-correlation shot noise as a function of the gate voltage V_g (solid line, with phonon mode; dashed line, without phonon mode). (c) The cross-correlation shot noise S_{LR}/w vs the frequency w for different coupling constants $g=0$ (solid curve), $g=0.3^2$ (dotted curve), $g=0.5^2$ (dashed curve), and $g=0.7^2$ (dashed-dotted curve), with $\Gamma=0.04$, $V_g=0$, $r=0.08$, and $\theta=88°$.

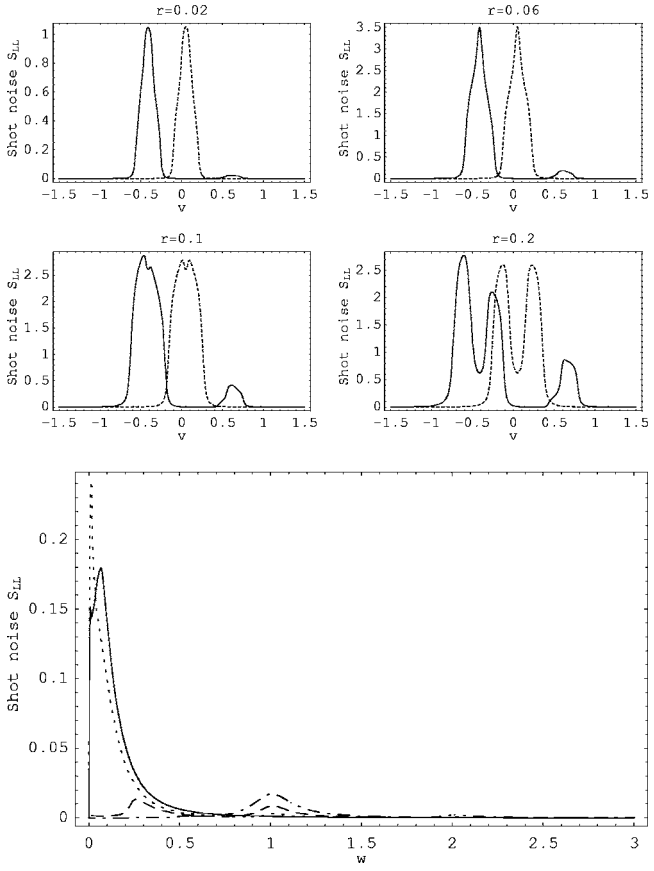

FIG. 4. (a) The autocorrelation shot noise vs the gate voltage V_g, with $r=0.02$, 0.06, 0.1, and 0.2 (solid line, with phonon mode, dashed line, without phonon mode). (b) The autocorrelation shot noise vs the frequency w with the same parameters as in Fig. 2(c) for various coupling constants $g=0$ (solid curve), $g=0.3^2$ (dotted curve), $g=0.5^2$ (dashed curve), and $g=0.7^2$ (dashed-dotted curve).

noise spectra exhibits two extra peaks, located symmetrically around the position of the main peak, and in the case of stronger magnetic field, the cross-correlation shot noise turns to negative for some gate voltages, while it remains positive for the other gate voltages. In Fig. 3(c), the oscillating behavior of the shot noise between positive and negative values can be observed and is due to the photon-assisted process.

We finally plot the autocorrelation shot noise vs the gate voltage V_g [Fig. 4(a)] and frequency w [Fig. 4(b)]. The autocorrelation shot noise spectra are also shifted, and a satellite peak appears, compared with the absence of the E-PH interaction. Moreover, the autocorrelation of the spin current is surely positive definite, which is different from the situation for cross-correlation. We see that the shot noises possess information about the phonon effect on the quantum transport through molecular devices.

IV. CONCLUSIONS

In summary, we have shown that the shot noise of the spin current in a single-molecule quantum dot can be significantly affected by the phonon mode and can provide beneficial information to improve the understanding of transport properties through the molecular QDs. It is shown that in addition to the shift of the resonant-peak position associated with the level of the dot, satellite peaks emerge at the integer number of the phonon frequency. The E-PH coupling can even reverse the sign of the zero-frequency cross-correlation shot noise.

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[41] The z component of field $B(t)$ splits the level ϵ into two levels, with $\epsilon_\downarrow < \epsilon_\uparrow$. The chemical potentials μ of the leads are adjusted by the gate voltage, so that $\epsilon_\downarrow < \mu < \epsilon_\uparrow$ and no bias voltage is applied to the two leads. A spin-down electron can tunnel into state $\epsilon_\downarrow$ from the left lead and absorb a photon to flip its spin and occupy the state $\epsilon_\uparrow$. Because $\epsilon_\uparrow > \mu$, it is easier for the spin-up electron to tunnel out to the leads. The same process occurs for spin-down electrons coming from the right lead. Therefore, spin-down electrons flow toward to the quantum dot, while spin-up electrons flow away from the dot, giving rise to a zero charge current and a net spin current.

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