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Interference effects on vibration-mediated tunneling through interacting degenerate molecular states

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Abstract

We study the combined effects of quantum electronic interference and Coulomb interaction on electron transport through near-degenerate molecular states with strong electron–vibration interaction. It is found that quantum electronic interference strongly affects the current and its noise properties. In particular, destructive interference induces pronounced negative differential conductances (NDCs) accompanying the vibrational excited states, and such NDC characters are not related to asymmetric tunnel coupling and are robust to the damping of a thermal bath. In a certain transport regime, the non-equilibrium vibration distribution even shows a peculiar sub-Poissonian behavior, which is enhanced by quantum electronic interference.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

In recent years, molecular electronics have been attracting tremendous interest for fertile nanoscale non-equilibrium phenomena induced by various interplays of interference, Coulomb and electron–vibration interactions for instance [1, 2]. Experimentally, molecules of small size are always weakly contacted to electrodes and the charging of the molecular states is often accompanied by notable mechanical motion. In these cases, the correlations between electron transfer events and the generations of correlated vibrations in molecules become so important that nonperturbative treatment of all the interactions within the molecules is essential. From this point of view, master equation approaches are widely used in theoretical studies on electron transport properties of molecules so that all these higher order correlations are included and only the weak contact couplings are treated perturbatively [3–7].

Transports through a single electronic state have been investigated theoretically in detail; however, situations with multiply electronic states are little studied within the master equation approach [8–12]; in particular, the role of electronic coherence, which is absent in the former cases, is not well understood. Recently, several theoretical groups found notable suppression of the current and pronounced NDC as the signature of interference in quantum dot systems without vibration, when the tunneling induced splitting and the coherent on site dynamics become of the order of the electron tunneling rate in the high temperature sequential tunneling regime [13–15]. This implies that transport measurements on near-degenerate electronic levels may exhibit visible interference effects even in current–voltage characteristics. Meanwhile, recent experiments on vibration-assisted tunneling in a suspended single-walled carbon nanotube (CNT) quantum dot suggested fourfold degenerate discrete electronic levels (charge and spin) in the Coulomb blockade regime [16–18]. Thus it is interesting to see how interference effects influence the transport properties through interacting near-degenerate electronic levels in the presence of vibration.

In this paper, we adopt a relatively simple model to investigate non-trivial physics solely due to quantum interference. Our spinless model consists of two interacting near-degenerate electronic orbitals both strongly coupled to a single vibrational mode. A Markovian master equation for sequential tunneling under the singular coupling limit is employed to explore the properties of electron and vibration

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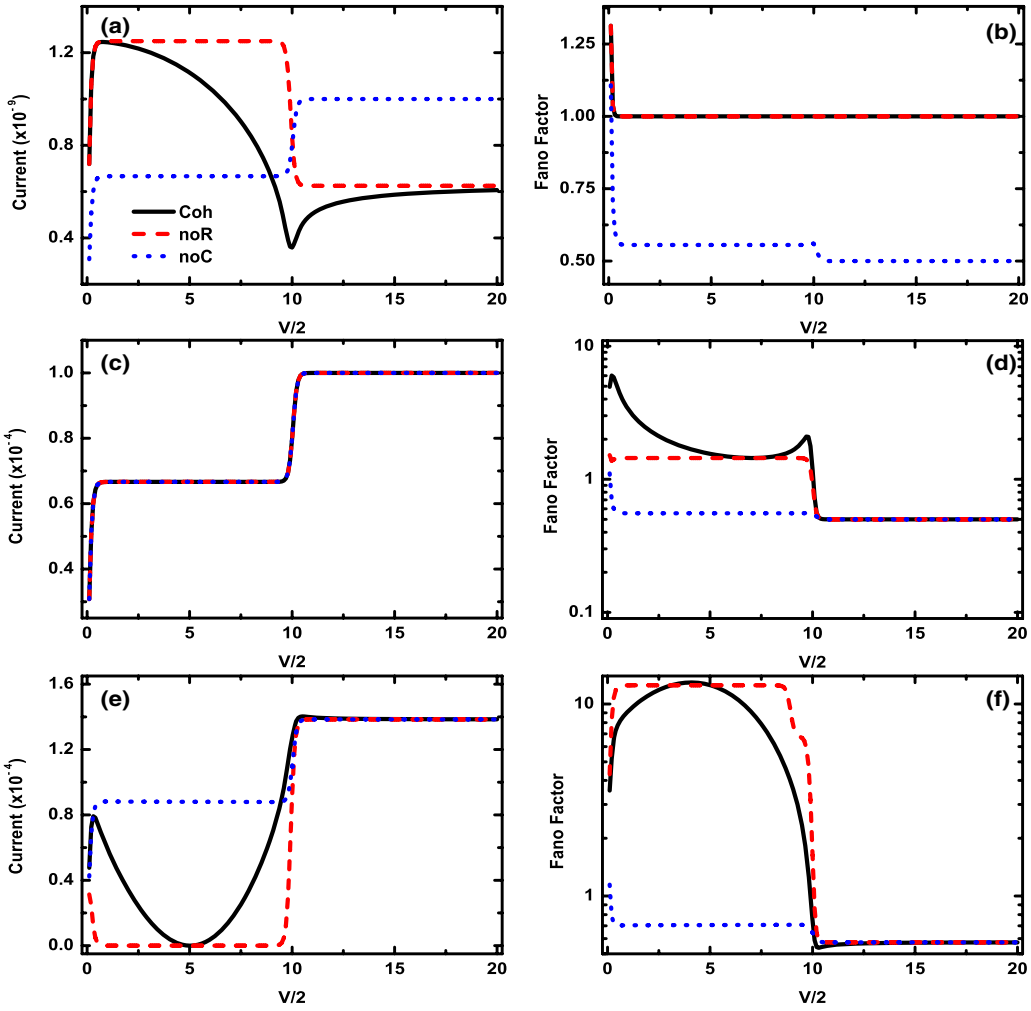

Figure 1. Current and the corresponding electronic Fano factor versus bias voltage for destructive interference (upper panels) with $t_{\rm L}^1 = -t_{\rm L}^2 = t_{\rm R}^1 = t_{\rm R}^2 = 10^{-2}$, complete constructive interference (middle panels) with $t_{\rm L}^1 = t_{\rm L}^2 = t_{\rm R}^1 = t_{\rm R}^2 = 10^{-2}$, and partial constructive interference (lower panels) with $t_{\rm L}^1 = 1.5t_{\rm L}^2 = 1.5t_{\rm R}^1 = t_{\rm R}^2 = 1.5 \times 10^{-2}$. The other parameters are $\epsilon = 0$, $u = 10$, $k_{\rm B}T = 0.1$ and $\Delta = 5 \times 10^{-7}$ except that for complete constructive interference $\Delta = 1 \times 10^{-4}$. Note that the values depicted by the dotted line in (a) are scaled by 1×10^{-5}, suggesting that the current is strongly suppressed by destructive interference.

dynamics, such as the current–voltage characteristics, the internal vibration distribution and their statistics described by electronic and vibrational Fano factors, respectively. Interestingly, it is found that almost every vibrational excitation accompanied by an NDC character is the consequence of destructive interference and it is robust to the damping of thermal bath and is not related to asymmetric tunnel coupling, which are in qualitative agreement with the results of a recent experiment about suspended CNT quantum dot [18].

2. Molecular model and master equation formalism

We consider a molecular model with two near-degenerate electronic levels coupled to a single local vibration mode ω, whose Hamiltonian reads ($\hbar = e = 1$)

$$H_{\rm mol} = \sum_{i=1,2} \epsilon_i n_i + u n_1 n_2 + \omega b^\dagger b + \omega \sum_{i=1,2} \lambda_i n_i (b^\dagger + b) \quad (1)$$

where $n_i = a_i^\dagger a_i$ is the electron number operator of level i and $b^\dagger(b)$ is the vibration creation (destruction) operator. u and λ represent Coulomb interaction energy and electron–vibration coupling respectively. Electrons in the molecule can be exchanged with two leads under symmetrically applied bias voltage via tunneling couplings and vibrations are damped by a thermal bath. The total Hamiltonian reads $H = H_{\rm mol} + H_{\rm L} + H_{\rm R} + H_{\rm B} + H_{\rm T} + H_{mb}$. The lead $H_r (r = {\rm L}, {\rm R})$ is modeled by non-interacting electrons with density of states ϱ_r, and the coupling to the leads is described by the tunneling terms $H_{\rm T}$ with lead- and orbital-dependent amplitudes t_r^i. Quantum interference effects which account for the role of the relative phase difference between two transmission paths, i.e. level 1 and level 2 in our model, can be included in a phenomenological way by assuming $t_{\rm L}^1 = |t_{\rm L}^1|$, $t_{\rm R}^{1(2)} = |t_{\rm R}^{1(2)}|$ and $t_{\rm L}^2 = \eta |t_{\rm L}^2|$, where $\eta = 1$ for constructive interference and $\eta = -1$ for destructive interference. In order to get maximal quantum interference effects, the energy-independent tunneling rates must be equal, i.e., $|\Gamma_{\rm L(R)}^1| = |\Gamma_{\rm L(R)}^2|$, where $\Gamma_r^i = 2\pi \varrho_r |t_r^i|^2$, implying complete constructive (destructive) interference for $\eta = 1(-1)$. The effect of the thermal bath

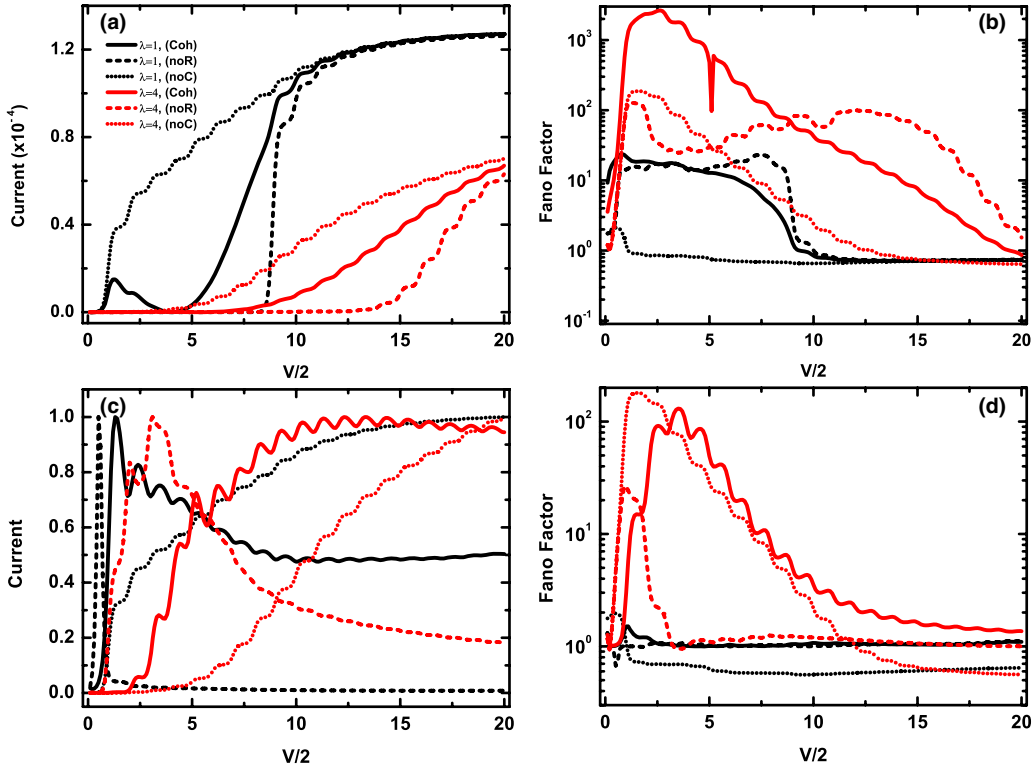

Figure 2. Current and the corresponding electronic Fano factor versus bias voltage with intermediate ($\lambda = 1$) and strong ($\lambda = 4$) electron–vibration couplings for partial constructive interference (upper panels) with $t_{\rm L}^1 = 1.5t_{\rm L}^2 = 1.5t_{\rm R}^1 = t_{\rm R}^2 = 1.5 \times 10^{-2}\omega$ and for destructive interference (lower panels) with $t_{\rm L}^1 = -t_{\rm L}^2 = t_{\rm R}^1 = t_{\rm R}^2 = 10^{-2}\omega$, respectively. The other parameters are $k_{\rm B}T = 0.1\omega$, $\tilde{u} = 10\omega$, $\tilde{\Delta} = 5 \times 10^{-7}\omega$ and $\tilde{\epsilon} = -\omega$ ($\tilde{\epsilon} = 0$) for intermediate (strong) electron–vibration coupling. The current values in (c) are normalized by their respective maximums.

$H_{\rm B} = \sum_l \omega_l b_l^\dagger b_l$ is also included in our model and the weak coupling between local vibrations and the thermal bath is described by $H_{mb} = (b^\dagger + b)\sum_l \kappa_l (b_l^\dagger + b_l)$.

The strong electron–vibration coupling can be eliminated via Lang–Firsov unitary transformation $U = \prod_{i=1,2} X_i^{n_i}$ with $X_i = {\rm e}^{-\lambda_i(b^\dagger - b)}$ [19], which renormalizes the Hamiltonian as

$$\tilde{H}_{\rm mol} = \tilde{\epsilon}(n_1 + n_2) + \tilde{\Delta}/2(n_2 - n_1) + \tilde{u}n_1 n_2 + \omega b^\dagger b \quad (2)$$

$$\tilde{H}_{\rm T} = \sum_{kri} (t_r^i X_i c_{kr}^\dagger a_i + {\rm h.c.}) \quad (3)$$

$$\tilde{H}_{mb} = \left(b^\dagger + b - 2\sum_{i=1,2} \lambda_i n_i\right) \sum_l \kappa_l (b_l^\dagger + b_l) \quad (4)$$

where the renormalized on site average energy $\tilde{\epsilon} = 1/2\sum_{i=1,2}(\epsilon_i - \omega\lambda_i^2)$, the renormalized electronic splitting $\tilde{\Delta} = \epsilon_2 - \epsilon_1 + \omega(\lambda_1^2 - \lambda_2^2)$ and the renormalized Coulomb energy $\tilde{u} = u - 2\lambda_1\lambda_2\omega$.

While this transformation facilitates a nonperturbative description of vibrational and electronic interactions, the molecule–lead and vibration–thermal bath couplings are treated within a second order perturbation master equation approach. When the high temperature regime ($k_{\rm b}T > \Gamma$) is implied, it is proper to only include sequential tunneling events and neglect the co-tunneling contributions and the effects of electronic level broadening due to tunnel coupling with leads. In this study, we are interested in a regime where the coherent on site dynamics, the electron tunneling rate and the tunneling induced level splitting are of the order of Γ. In this regime the temperature of the leads is too high to resolve the energy difference $\tilde{\Delta}$ by average current measurements by scanning the voltage of the leads. As suggested in [20], the singular coupling limit can properly describe the competition between the coherent dynamics in the molecule with near-degenerate orbitals and the electron transport due to tunneling. In this limit, we rescale $t_r^i \to \gamma t_r^i$, $\kappa_l \to \gamma\kappa_l$ and $\tilde{\Delta} \to \gamma^2\tilde{\Delta}$ so that the ratio $\Gamma/\tilde{\Delta}$ remains constant when $\gamma \to 0$.

All observables of interest, such as the current–voltage characteristics and the vibrational excitation $\langle b^\dagger b\rangle$, can be obtained from the reduced density matrix ρ of the electronic and vibrational degrees of freedom of the molecule within the Markovian master equation approach (letting $\gamma \to 0$),

$$\frac{\rm d}{{\rm d}t}\rho(t) = -{\rm i}[H_{\rm mol}^-, \rho(t)] - \int_0^\infty {\rm d}s\, {\rm Tr}_E(L_E {\rm e}^{-{\rm i}L_{\rm mol}^+ s} L_E \rho_{\rm leads}^0 \rho_{\rm B}^0)\cdot\rho(t) \quad (5)$$

where $H_{\rm mol}^- = \tilde{\Delta}/2(n_2 - n_1)$, dissipative superoperator $L_E = [\tilde{H}_{\rm T} + \tilde{H}_{mb}, \cdot]$ and $L_{\rm mol}^+ = [(\tilde{H}_{\rm mol} - H_{\rm mol}^-), \cdot]$. $\rho_{\rm leads}^0$ and $\rho_{\rm B}^0$ denote the equilibrium density matrix of the leads and the thermal bath respectively. Note that the electronic splitting $\tilde{\Delta}$ does not appear in the dissipative term but is retained in the free evolution part, which implies we treat the tunneling induced

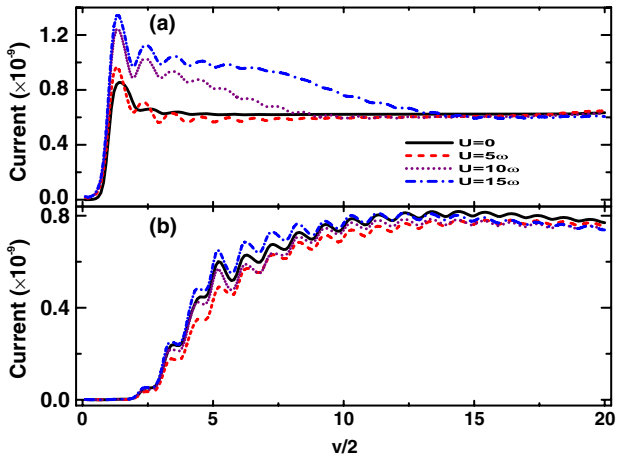

Figure 3. Current versus bias voltage for destructive interference with intermediate (a) ($\lambda = 1$) and strong (b) ($\lambda = 4$) electron–vibration couplings for different values of the renormalized Coulomb repulsive energy U. The other parameters are as in figure 2(c).

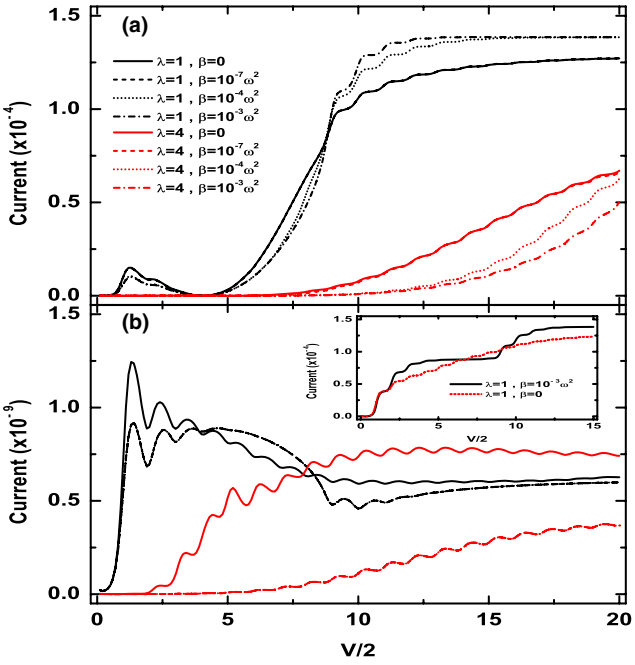

Figure 4. Current versus bias voltage for partial constructive interference (a) and destructive interference (b) for different values of the vibration damping rate β. The other parameters of (a)/(b) are as for the case of keeping full coherence (Coh) in figure 2(a)/(b). The inset in (b) plots the incoherent (noC) current with intermediate ($\lambda = 1$) electron–vibration coupling for unequilibrated ($\beta = 0$) and almost equilibrated ($\beta = 10^{-3}\omega^2$) vibrations.

level renormalization and any other level splitting on the same order in the singular coupling limit.

We rewrite equation (5) in a compact form [21]: $\frac{\mathrm{d}}{\mathrm{d}t}\rho(t) = -\mathrm{i}[H_{\mathrm{mol}}^{-}, \rho(t)] + \sum_{r=\mathrm{L,R}}(-\Pi_r + \Sigma_r^{+} + \Sigma_r^{-})\rho(t) + L_p\rho(t)$. The dissipative term is separated into the diagonal contribution Π_r that leaves the number of electrons in the molecule unchanged, two off-diagonal parts Σ_r^{+} and Σ_r^{-} for increase and decrease in the number of electrons, respectively, and the thermal bath contribution L_p for damping vibrations in the molecule. From the point of view of counting statistics, this separation is essential to keep track of the trajectories of single electron transfers and to calculate the transfer probability $P(k,t)$ of k electrons in the time interval t through one lead. Generally, we obtain first the generating function (GF), which is defined as $G(\chi,t) = \sum_k \mathrm{e}^{\mathrm{i}k\chi}P(k,t)$, where χ is the counting field at one lead. From the GF, we get the current $I = -\frac{\mathrm{i}}{t}\partial_\chi \ln G(\chi,t)|\chi \to 0, t \to \infty$ and the shot noise $S(0) = -\frac{1}{t}\partial_\chi^2 \ln G(\chi,t)|_{\chi\to 0, t\to\infty}$ (Fano factor $F = S(0)/I$). The detailed calculations of the GF can be found in [21].

In contrast to the usual rate equation method, we fully take into account the electronic coherences of the density matrix ρ, while vibrational coherences are negligible since we consider the regime $\omega > k_{\mathrm{b}}T$ with visible quantization effects of the vibrational mode. In practical calculation, we project equation (5) on the many-body eigenstates $|\upsilon_m, n\rangle$ (vibration number n and electronic state $|\upsilon_m\rangle$ with electron number m) of the renormalized molecular Hamiltonian. A total of 60 vibrational states are included in our calculation. This number is enough according to the vibration quanta and the bias we used. The time integrations as shown in equation (5) can be performed by using the relationship $\int_0^\infty \mathrm{d}s\, \mathrm{e}^{\mathrm{i}\omega s} = \pi\delta(\omega) + \mathrm{i}\mathcal{P}\frac{1}{\omega}$, which results in two terms describing the real electron transfer shifting the number of electrons on the molecule and the tunneling induced level renormalization effects (or so-called virtual electron transfer) changing the molecular states singly occupied by an electron. As we will see below, such renormalizations play an important role in electron transport through near-degenerate levels with small bias voltage.

3. Results and discussion

3.1. NDC mechanisms in a vibration-free molecule due to quantum interference

Firstly, we point out some major results of electron transport through a molecule without vibrational degrees of freedom, which has been investigated theoretically with different approaches under various approximations [13–15]. Figure 1 shows the current–voltage and Fano factor characteristics in the high temperature regime ($k_{\mathrm{b}}T \gg \Gamma \geqslant \tilde{\Delta}$) based on three different approximations as keeping full coherence (Coh), neglecting level renormalizations (noR) and neglecting all coherence (noC). It is interesting to note that destructive interference induces strongly NDC phenomena shown in the current–voltage characteristic in addition to obvious suppression of electron tunneling. The current drop around $eV_{\mathrm{bias}} \approx 2u$ (see the dashed line in figure 1(a)) where the double occupied state enters the transport window is caused by different decoherence rates due to electron tunneling for singly and double occupied states [14], while the NDC feature in the low bias regime ($eV_{\mathrm{bias}} < 2u$), where the strong Coulomb interaction prohibits the formation of the double occupied state, is due to bias-dependent level renormalizations [13], as shown by the solid line in figure 1(a). In contrast, the current–voltage characteristic does not manifest the signature

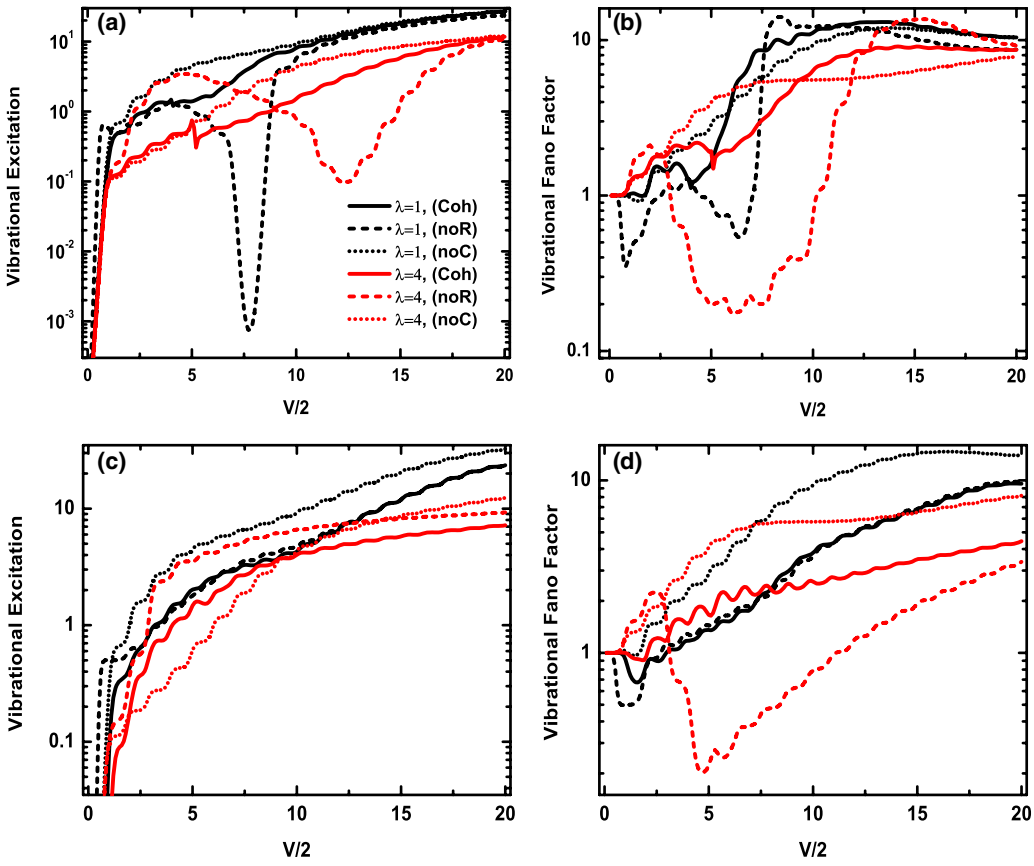

Figure 5. Vibrational excitation and the corresponding vibrational Fano factor versus bias voltage with intermediate ($\lambda = 1$) and strong ($\lambda = 4$) electron–vibration couplings for partial constructive interference (upper panels) and for destructive interference (lower panels). All the parameters are as in figure 2.

of complete constructive interference. However, the strong super-Poissonian feature in the low bias regime suggests the role of constructive interference, which enables one to decouple one of the levels from both left and right leads by a unitary transformation of the molecular Hamiltonian as long as $|\Gamma^1_{\mathrm{L(R)}}| = |\Gamma^2_{\mathrm{L(R)}}|$. In this case electrons may be trapped by the isolated level for a long time when transported through the molecule so that electrons tend to be transferred in bunches, resulting in the super-Poissonian behavior. If $|\Gamma^1_{\mathrm{L(R)}}| \neq |\Gamma^2_{\mathrm{L(R)}}|$, such a trapping effect can be loosened and a mechanism of slow and fast transport channels caused by level renormalizations comes into play so that a remarkable NDC character can be found in the low bias regime due to this partial constructive interference [20]. However, we have to recall that all such quantum interference effects on the current–voltage and Fano factor characteristics become invisible when the energy splitting Δ becomes larger than Γ.

3.2. Electronic interference effects on vibration-mediated electron tunneling

Next we turn to study the case with intermediate ($\lambda_1 = \lambda_2 = 1$) and strong ($\lambda_1 = \lambda_2 = 4$) electron–vibration couplings in the absence of vibration damping. As we are only concerned with the role of quantum interference in the presence of vibration, combined effects with intramolecular asymmetry ($\lambda_1 \neq \lambda_2$) [8, 11] are out of our consideration. In the following calculations, we set $k_{\mathrm{b}}T = 0.1\omega$ and keep $\tilde{u} = 10\omega$ for two electron–vibration couplings. The comparison of results due to partial constructive and complete destructive interference effects reveals their different role in vibration-mediated tunneling properties as shown in figure 2. For the partial constructive one on the one hand, the current is systematically rectified, as compared to the vibration-free case, with the characteristic Franck–Condon steps indicating molecular states with increasing number of vibration available for electron transport by increasing the bias voltage. However, while a pronounced NDC is found for the case of keeping full coherence and $\lambda = 1$ in the low bias regime, the current for $\lambda = 4$ is strongly suppressed so that NDC is invisible compared with the large scale of its high bias current. Such current suppression is a signature of the so-called Franck–Condon blockade arising from vibration-mediated tunneling couplings which induce exponentially suppressed electron transitions between low-lying vibrational states. This blockade effect also manifests itself in huge zero frequency electronic Fano factors as shown in figure 2(b), and the electronic Fano factor for the coherent one being much larger than that for no coherence shows that constructive interference makes this blockade mechanism more effective.

For destructive interference on the other hand, there are always strong NDCs accompanying the vibrational excited

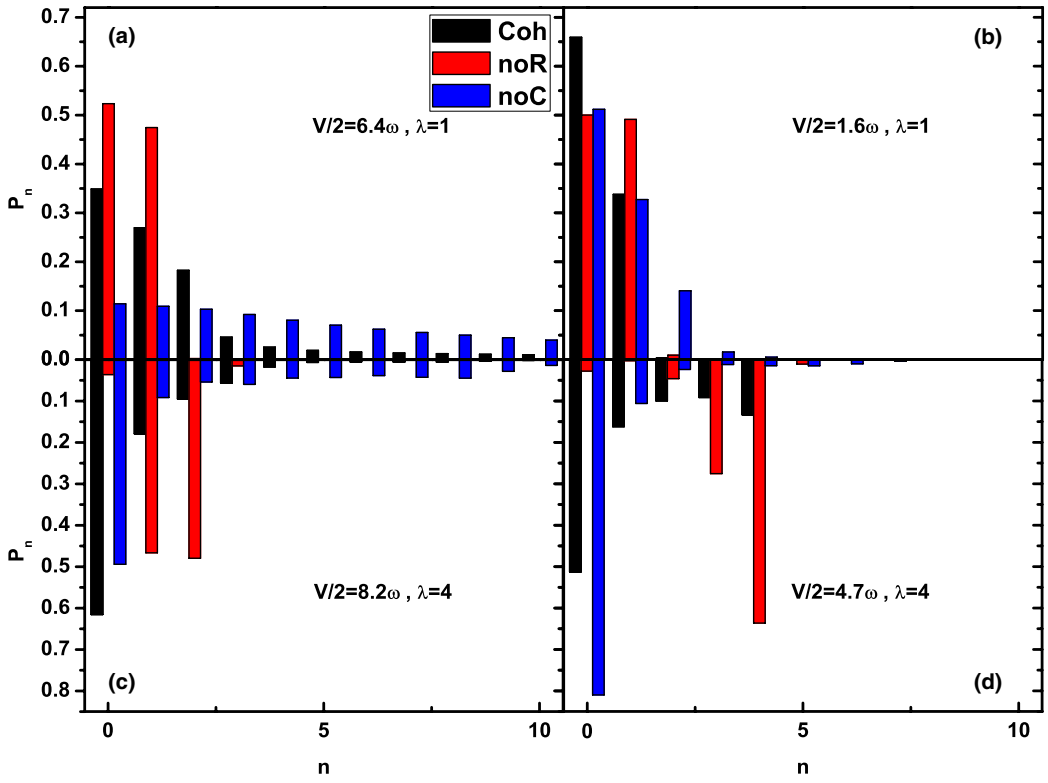

Figure 6. Vibration distributions P_n as a function of vibration number n for partial constructive interference (left panels) and for destructive interference (right panels) at different bias voltages where obvious sub-Poissonian features are shown. All the parameters of (a) and (c) are as in figure 5(b) and the ones of (b) and (d) are as in figure 5(d).

states which become available for electron transport. As shown by the dashed line in figure 2(c), it is much more obvious when neglecting the renormalizations that the current shows distinct step-like monotonic decrease characteristics. Such current drops can be explained by different decoherence rates for molecular states of different number of vibration. However, the bias-dependent renormalizations induce oscillating-like decrease (increase) behavior in current–voltage characteristic for intermediate coupling $\lambda = 1$ (strong coupling $\lambda = 4$) when a certain vibrational excited state comes into play. The oscillating-like increase trend of current for strong coupling can be consider as the result of Franck–Condon blockade, which also contributes to the huge Fano factor. In contrast to the partial constructive one, the NDCs are visible not only in the low bias regime but also in the large bias regime, where double electron occupied states are open for transport, which implies that a finite renormalized Coulomb repulsive energy ($\tilde{u} > 0$) is not so vital for the destructive interference to produce NDCs as for the partial constructive one. Figure 3 shows the results for different values of $\tilde{u}$ for destructive interference and we find that strong NDCs even exist when $\tilde{u} = 0$. This fact indicates the role of bias-dependent renormalizations, resulting from electron tunneling through different vibrational states, in contrast to the vibrational-free case where such renormalizations are bias independent for $u = 0$. Additionally, we also find that these pronounced NDCs are slightly affected by changing the asymmetric coupling to the leads (not shown), which is consistent with the result of a recent experiment on suspended CNT quantum dots with multiple fourfold degeneracies (spin and valley) of electronic states [18]. This suggests that the valley degeneracy in such a clean carbon nanotube may exhibit destructive interference features.

Now we discuss the effect of damping due to an additional thermal bath, whose contribution is written $L_p\rho = \frac{\beta}{2}[-b^\dagger b\rho + 2b\rho b^\dagger - \rho b^\dagger b] + \eta\beta[-b^\dagger b\rho + b\rho b^\dagger + b^\dagger\rho b - \rho b^\dagger b]$, where $\eta = 1/(\mathrm{e}^{\omega/k_\mathrm{b}T} - 1)$ and the decay rate of vibrational quanta into the thermal bath is β. We assume that the temperature of the thermal bath is the same as the temperature of the leads. In figure 4 it is shown that although the current is more sensitive to the damping for destructive interference than for partial constructive interference, one can still find notable NDCs accompanying each vibrational excitation. However, resonant absorption processes are not activated because of strong damping, which is most obvious when coherence is completely neglected (see the inset in figure 4(b)).

3.3. Electronic interference effects on internal vibration generation

Since vibrations on the molecule are strongly correlated with electron current, electron interference may also have an impact on the distribution of non-equilibrium vibrational excitations and its statistics, which is characterized by the vibrational Fano factor $F_\mathrm{vib} = \frac{\langle n^2\rangle - \langle n\rangle^2}{\langle n\rangle}$ (vibration number operator $n = b^\dagger b$). Compared with the model of a single electronic level, there are additional resonant vibrational quanta absorption

processes [12] associated with the sequential tunneling of two electrons from the source lead at low bias voltage $V < 2\tilde{u}$. Such resonant absorption processes provide a mechanism to stabilize single-molecule junctions, which is much obvious in the presence of electronic coherences. The results of neglecting renormalizations for partial constructive interference (see figure 5(a)) even show a pronounced reduction of vibrational excitations while increasing the bias. This can be explained in terms of the vibrational energy excited by a tunneling electron which is finally trapped on a level almost decoupled from the drain lead enabling electron transport through the molecule from another level against the Coulomb repulsion via such resonant absorption processes and thus the molecule has much more chance to be cooled while increasing the bias.

Generally, one always find a super-Poissonian character for an equilibrium bosonic bath and it is of particular interest to seek possible non-classical features in non-equilibrium boson distributions. In some transport regimes of a quantum dot which is coupled to a harmonic oscillator, the distribution of vibration number shows a sub-Poissonian behavior with small electron–vibration coupling $\lambda < 1$ and asymmetric tunnel couplings [22]. As to our model of near-degenerate molecular states, one can find such sub-Poissonian character even for strong electron–vibration coupling $\lambda \geqslant 1$ and symmetric tunnel couplings as long as one takes the interference effect into account. This peculiar feature is most remarkable when renormalizations are neglected as shown in figures 5(b) and (d). This sub-Poissonian behavior originates from a particular vibration distribution, so-called selective population of vibration states [22], where the occupation probabilities of some excited vibration states are comparable to or even larger than the one of the ground states (see figure 6). In this situation the vibration number is increased or decreased by certain numbers accidentally when electron tunneling takes place.

4. Conclusions

In summary, we have used a Markovian master equation to address the role of quantum interference in the non-equilibrium electron transport and vibration distribution properties of a spinless molecular junction, consisting of two near-degenerate electronic states strongly coupled to a single vibrational mode. The singular coupling limit is applied to properly treat the competition between the dynamics of many-body coherence (only electronic coherence is considered here) and electron sequential tunneling at high temperature. While partial constructive interference only induces a visible NDC structure in the low bias regime for small or intermediate electron–vibration coupling, a series of robust NDCs is found to accompany almost each vibrational excitation as a consequence of destructive interference. Additionally, quantum interference even enhances the sub-Poissonian feature of vibration distribution in a certain transport regime.

Acknowledgments

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