

# Charge transfer statistics of a molecular quantum dot with a vibrational degree of freedom

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We analyze the full counting statistics of a single-site quantum dot coupled to a local Holstein phonon for arbitrary transmission and weak electron-phonon coupling. We identify explicitly the contributions due to quasielastic and inelastic transport processes in the cumulant generating function and discuss their influence on the transport properties of the dot. We find that in the low-energy sector, i.e., for bias voltage and phonon frequency much smaller than the dot-electrode contact transparency, the inelastic term causes a sign change in the shot noise correction at certain universal values of the transmission. Furthermore, we show that when the correction to the current due to inelastic processes vanishes, all the odd order cumulants vanish as well.

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During the past decade molecular electronics has evolved into an important branch of condensed-matter physics.<sup>1</sup> Nowadays it is possible to electrically contact molecules of almost any geometry and complexity ranging from carbon nanotubes to hydrogen molecules.<sup>2–7</sup> Moreover, the experimental observables are not restricted to the linear conductance properties anymore but encompass also the nonlinear current-voltage characteristic  $I(V)$  as well as the shot noise.<sup>5</sup> Especially in the case when vibrational degrees of freedom are involved, these transport quantities display a number of very interesting features.<sup>8–15</sup>

One of the most pronounced effects of the interaction between the electronic and vibronic degrees of freedom is the abrupt change in the system conductance once the applied voltage is increased beyond a threshold, which is related to the excitation energy of molecular vibrations.<sup>3,4,8,12,15–19</sup> This is the reason why measurements around this turning point have become an invaluable instrument for the experimental investigation of such systems. Interestingly, the sign of this conductance step depends crucially on the junction transparency, being negative for almost perfect transmission and positive in the opposite case. Most theoretical efforts have been centered around these two limiting cases.<sup>9,16,17,19–23</sup> According to Ref. 19, the transition is nonuniversal and the precise condition involves all system parameters.<sup>24</sup> In general it occurs at an intermediate value of the transmission.

Thus far mainly the nonlinear current-voltage characteristic has been analyzed in this regime. However, for future applications it is of importance to also possess information about the noise properties of such systems.<sup>5,20</sup> A very convenient tool to calculate a variety of transport properties is the full counting statistics (FCS), which gives the probability distribution  $P(Q)$  to transfer  $Q$  elementary charges during a fixed (very long) waiting time  $T$ . The average value  $\langle\langle Q \rangle\rangle$  is then directly related to the current and its variance  $\langle\langle Q^2 \rangle\rangle$  to the noise power. The relevance of higher-order cumulants  $\langle\langle Q^n \rangle\rangle$  has been demonstrated in Ref. 25. Moreover, the analytic structure of the cumulant generating function  $\ln \chi(\lambda)$  can give invaluable insights into the nature of the processes contributing to the transport.<sup>26–30</sup> That is the reason why we would like to address the FCS of the molecular quantum dot coupled to a Holstein phonon.

We model the system by the following Hamiltonian:

$$H = H_L + H_R + H_d + H_T + H_{ph} + H_{el,ph}. \quad (1)$$

The terms  $H_{L,R}$  describe the left and right electrodes in the language of the respective electron field operators  $\psi_{L,R}(x)$ . We model them as noninteracting fermionic continua held at the chemical potentials  $\mu_{L,R}$ . The applied bias voltage is then given by  $V = \mu_L - \mu_R > 0$  (we use units where  $e = \hbar = 1$ ). The particle transport between the dot and the electrodes is mediated by a local (symmetric<sup>31</sup>) tunneling coupling at  $x=0$  with an amplitude  $\gamma$ ,

$$H_T = \gamma d^\dagger [\psi_L(x=0) + \psi_R(x=0)] + \text{H.c.}, \quad (2)$$

where  $d$  is the annihilation operator of the electron on the dot. We assume it to be spinless and modeled by  $H_d = \Delta d^\dagger d$ , with  $\Delta$  being the bare energy of the dot state, which can be tuned by an applied gate voltage. The local (single-mode) phonon is described by  $H_{ph} = \Omega a^\dagger a$  while the electron-phonon coupling term with the amplitude  $g$  is given by

$$H_{el,ph} = g q d^\dagger d, \quad (3)$$

where  $q = a + a^\dagger$  is proportional to the phonon displacement operator.

In order to determine the FCS, we calculate the cumulant generating function (CGF)  $\ln \chi(\lambda) = \ln \langle e^{i\lambda Q} \rangle$  which gives access to the cumulants (or irreducible moments)  $\langle\langle Q^n \rangle\rangle$  of  $P(Q)$ . The formalism for CGF calculation has been developed in Refs. 32 and 33 and is by now well adapted to quantum impurity problems.<sup>30,34,35</sup> We chose to use perturbation theory in the electron-phonon coupling  $g$ . The corresponding CGF then reads as

$$\ln \chi(\lambda) = \ln \chi_0(\lambda) + \ln \chi'(\lambda). \quad (4)$$

The first term is the CGF of the clean system at  $g=0$ . At zero temperature, it is known to be given by

$$\ln \chi_0(\lambda) = T \int_{-V/2}^{V/2} \frac{d\omega}{2\pi} \ln [1 + T_0(\omega)(e^{i\lambda} - 1)], \quad (5)$$

where  $T_0(\omega) = \Gamma^2 / [(\omega - \Delta)^2 + \Gamma^2]$  is the single-particle transmission coefficient of the resonant-level model<sup>30,36</sup> and  $\Gamma = 2\pi\rho_0\gamma^2$  is the dot-electrode contact transparency, which in the wide flat band model depends on the energy-independent density of electronic states in the electrodes  $\rho_0$ . The correction is treated in the spirit of Ref. 37, which is based on the

generalized Keldysh approach proposed in Ref. 26. It is given by

$$\chi'(\lambda) = \langle T_C e^{-ig \int_C ds q(s) d^\dagger(s) d(s)} \rangle_\lambda, \quad (6)$$

where the expectation value is taken with respect to the non-interacting Hamiltonian  $H_L + H_R + H_d + H_T^\lambda + H_{ph}$  and is time ordered on the Keldysh contour  $\mathcal{C}$ . The dependence on the counting field  $\lambda$  is contained in  $H_T^\lambda = \gamma d^\dagger (e^{i\lambda} \psi_L + \psi_R) + \text{H.c.}$  The counting field has different signs on the two Keldysh branches,  $\lambda(t) = \pm \lambda$  for  $t \in \mathcal{C}_\pm$ .

An application of the standard linked cluster expansion generates two different contributions to the lowest-order ( $g^2$ ) term:  $\ln \chi' = \ln \chi_1 + \ln \chi_2$ . The first one is present only if  $\Delta \neq 0$  and is thus a consequence of a detuning of the dot from the particle-hole symmetric point,

$$\ln \chi_1(\lambda) = -\frac{ig^2 T}{2} \sum_{k,l=\pm} (kl) \int ds_1 ds_2 A^{kl}(s_1 - s_2) N^k(s_1) N^l(s_2). \quad (7)$$

Here,  $A(t, t') = -i \langle T_C q(t) q(t') \rangle_0$  denotes the phonon Green's function (GF) in Keldysh space and  $N^k(s) = \langle d^\dagger(t_k) d(t_k) \rangle_\lambda$

$-1/2$  for  $t_k \in \mathcal{C}_k$ ;  $k = \pm$  are generalized dot population probabilities. Due to the explicit  $\lambda$  dependence on the Keldysh branch  $k$  they are different on the forward/backward path  $\mathcal{C}_k$ . The other contribution is given by

$$\ln \chi_2(\lambda) = -\frac{g^2 T}{2} \sum_{k,l=\pm} (kl) \int \frac{d\omega}{2\pi} \pi^{kl}(\omega) A^{lk}(\omega), \quad (8)$$

where the generalized charge polarization loops are defined in terms of the dot level GFs  $D(t) = -i \langle T_C d(t) d^\dagger(t') \rangle_\lambda$  by

$$\pi^{kl}(\omega) = i \int \frac{d\omega'}{2\pi} D^{kl}(\omega + \omega') D^{lk}(\omega'). \quad (9)$$

Both contributions (7) and (8) can be calculated using the unperturbed dot GFs,<sup>30</sup>

$$D(\omega) = \begin{pmatrix} D^{--} & D^{+-} \\ D^{+-} & D^{++} \end{pmatrix} = \frac{1}{\mathcal{D}_0(\omega)} \begin{pmatrix} (\omega - \Delta) + i\Gamma(n_L + n_R - 1) & i\Gamma(e^{i\lambda/2} n_L + e^{-i\lambda/2} n_R) \\ i\Gamma[e^{-i\lambda/2}(n_L - 1) + e^{i\lambda/2}(n_R - 1)] & -(\omega - \Delta) + i\Gamma(n_L + n_R - 1) \end{pmatrix}, \quad (10)$$

where the denominator is given by

$$\mathcal{D}_0(\omega) = (\omega - \Delta)^2 + \Gamma^2 [n_L(1 - n_R)(e^{i\lambda} - 1) + n_R(1 - n_L)(e^{-i\lambda} - 1) + 1]. \quad (11)$$

In the wide flat band model the electrodes are noninteracting fermions with Fermi distribution functions  $n_{R,L}(\omega)$ . The chemical potentials in the left/right electrode are  $\pm V/2$ . From now on we concentrate on the zero-temperature results. The presented results hold for all temperatures much smaller than the smallest energy scale in the system. The phonon GFs are given by  $A^{--(+)}(t) = -i \exp(\mp i\Omega|t|)$  and  $A^{++(-)}(t) = -i \exp(\pm i\Omega t)$ . Equation (10) allows one to calculate the generalized population probabilities  $N^k$  and one finds from Eq. (7)

$$\ln \chi_1(\lambda) = -\frac{g^2 T}{2\pi^2 \Omega} \ln \left[ \frac{f(V/2)}{f(-V/2)} \right] \sum_{p=\pm} p \arctan \left( \frac{V/2 - p\Delta}{\Gamma} \right), \quad (12)$$

where we defined a  $\lambda$ -dependent function  $f(\omega) = [1 + T_0(\omega)(e^{i\lambda} - 1)]^{-1}$ . The contribution  $\ln \chi_2$  can be written as a sum of two terms, which describe *quasielastic* and *inelastic* processes.<sup>19</sup> The quasielastic part reads as

$$\ln \chi_{qel}(\lambda) = -\frac{T}{2\pi} \int_{-V/2-\Delta}^{V/2-\Delta} d\omega \frac{\omega}{\omega^2 + \Gamma^2} e^{i\lambda} \Sigma_R^R(\omega), \quad (13)$$

where

$$\Sigma_R^R(\omega) = \sum_{k,l=\pm} \frac{g^2 \Gamma}{(\omega + k\Omega)^2 + \Gamma^2} \left\{ \frac{\omega + k\Omega}{2\Gamma} \left[ 1 + \frac{2k}{\pi} \arctan \left( \frac{lV/2 - \Delta}{\Gamma} \right) \right] + \frac{k}{\pi} \ln \left[ \frac{\sqrt{(lV/2 - \Delta)^2 + \Gamma^2}}{|\omega + k\Omega - (lV/2 - \Delta)|} \right] \right\} \quad (14)$$

is the real part of the retarded dot self-energy  $\Sigma^R(\omega)$ .<sup>19</sup> It is related to the formation of the phonon sidepeak in the spectral function of the dot and can be considered as a renormalization of the bare transmission.

The other contribution is the *inelastic* one.<sup>38</sup> It only contributes for  $V > \Omega$  and is given by

$$\ln \chi_{inel}(\lambda) = \frac{g^2 T}{2\pi} \theta(V - \Omega) \int_{-V/2-\Delta}^{V/2-\Delta} d\omega \sum_{k=\pm} \theta(V/2 - k\Delta - k\omega - \Omega) \frac{1}{\omega^2 + \Gamma^2} e^{i\lambda} \left[ \frac{\omega(\omega + k\Omega)}{(\omega + k\Omega)^2 + \Gamma^2} - \frac{1}{2} \frac{\omega(\omega + k\Omega) - \Gamma^2 e^{i\lambda}}{(\omega + k\Omega)^2 + \Gamma^2 e^{i\lambda}} \right]. \quad (15)$$

Equations (12), (13), and (15) are the main result of this Rapid Communication. Compatible results were obtained independently in two related studies.<sup>39,40</sup> They represent the

full perturbative result for the correction to the CGF due to the phonon at arbitrary transmission and are valid at zero temperature and for small  $g$ .<sup>41</sup>

The term  $\ln \chi_1$  is due to the renormalization of the level energy  $\Delta$  by the presence of the phonon<sup>19</sup> and is the leading term for small  $\Omega$ . This is precisely the condition when the adiabatic (or Born-Oppenheimer) approximation is valid. The interpretation of other contributions is more lucid in the low-energy sector when  $\Omega$ ,  $V \ll \Gamma, \Delta$ . Then we obtain

$$\ln \chi_1(\lambda) = \frac{2g^2 T \Delta V}{\pi^2 \Omega} \arctan\left(\frac{\Delta}{\Gamma}\right) \frac{1}{\Delta^2 + \Gamma^2 e^{i\lambda}}, \quad (16)$$

$$\ln \chi_{\text{qel}}(\lambda) = -\frac{g^2 T V}{\pi} \frac{\Delta^2}{\Delta^2 + \Gamma^2} \frac{1}{\Delta^2 + \Gamma^2 e^{i\lambda}}, \quad (17)$$

$$\begin{aligned} \ln \chi_{\text{inel}}(\lambda) &= \frac{g^2 T (V - \Omega)}{\pi} \theta(V - \Omega) \left[ \frac{\Delta^2}{\Delta^2 + \Gamma^2} - \frac{1}{2} \frac{\Delta^2 - \Gamma^2 e^{i\lambda}}{\Delta^2 + \Gamma^2 e^{i\lambda}} \right] \frac{1}{\Delta^2 + \Gamma^2 e^{i\lambda}}. \end{aligned} \quad (18)$$

In many cases the perturbative FCS of correlated systems is a linear combination of  $e^{in\lambda}$  terms with some integer  $n$ .<sup>26–29,42,43</sup> One possible explanation is that the corresponding terms describe transport processes in which the initial dot state is restored after the tunneling of  $n$  electrons. However,  $e^{i\lambda}$  enters the above equations *nonlinearly*. This means that there are processes in which  $n$  electrons are necessary in order to bring the system back to its initial state: the dot can be excited by a first transmitted electron and then a number of them can flow without interaction. The final  $n$ th electron then de-excites the system and leaves it in the initial ground state.

An expansion of Eqs. (16)–(18) in  $\Delta/\Gamma$ , which corresponds to the limit of large transmission  $T_0(\omega) \approx 1$ , leads to a series containing  $e^{-in\lambda}$  ( $n > 0$ ), describing electron *back-scattering* off the dot and thus a reduction in transmission. In the opposite limit of weak transmission,  $T_0(\omega) \approx 0$ , the CGFs can be expanded for small  $\Gamma/\Delta$ . One then only encounters *forward-scattering* terms containing  $e^{in\lambda}$  ( $n > 0$ ), which increase the transmission.

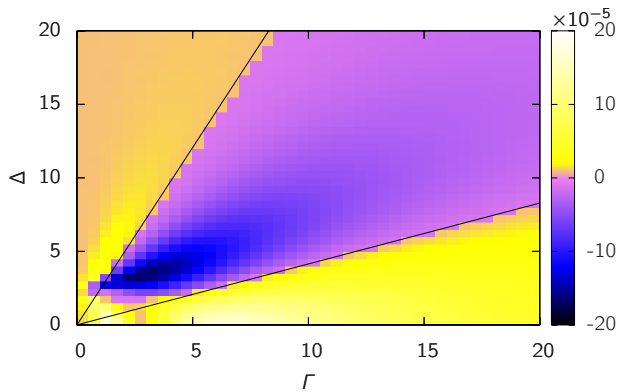


FIG. 1. (Color online) Density plot of the inelastic noise contribution  $\delta\langle Q^2 \rangle_{\text{inel}}/T$  for the parameters  $\Omega=10g$  and  $V=10.1g$  as a function of  $\Gamma$  and  $\Delta$  measured in units of  $g$ . Thin lines correspond to the transmissions given by Eq. (21). Dark and bright areas represent negative and positive noise correction values, respectively.

Next we would like to discuss the issue of the sign change of the conductance and noise corrections in vicinity of  $V = \Omega$ . In the low-energy sector it is only governed by the inelastic term. The correction to the current is given by

$$\begin{aligned} \delta I_{\text{inel}} &= -\frac{i}{T} \frac{d}{d\lambda} \bigg|_{\lambda=0} \ln \chi_{\text{inel}}(\lambda) \\ &= -\frac{g^2 (V - \Omega)}{2\pi} \theta(V - \Omega) \frac{\Gamma^2 (\Gamma^2 - \Delta^2)}{(\Gamma^2 + \Delta^2)^3}. \end{aligned} \quad (19)$$

Here we immediately realize that the sign change occurs at precisely  $\Delta = \pm \Gamma$ . As the low-energy transmission coefficient is given by  $T_0(0) = \Gamma^2 / (\Gamma^2 + \Delta^2)$ , the turning point condition is indeed  $T_0 = 1/2$ . Interestingly, not only the current but *all the odd order cumulants* (due to the inelastic correction) vanish at this point.

The correction to the noise power due to the inelastic tunneling is given by

$$\begin{aligned} \delta\langle Q^2 \rangle_{\text{inel}} &= -\frac{d^2}{d\lambda^2} \bigg|_{\lambda=0} \ln \chi_{\text{inel}}(\lambda) \\ &= \frac{g^2 T (V - \Omega)}{2\pi} \theta(V - \Omega) \frac{\Gamma^2 (\Gamma^4 - 6\Gamma^2 \Delta^2 + \Delta^4)}{(\Gamma^2 + \Delta^2)^4}. \end{aligned} \quad (20)$$

This term changes its sign at the values  $\Delta = (\pm 1 \pm \sqrt{2})\Gamma$ , which correspond to the bare transmission coefficients given by

$$T_0 = \frac{1}{2} \left( 1 \pm \frac{\sqrt{2}}{2} \right). \quad (21)$$

In Fig. 1, we have plotted the inelastic noise contribution resulting from CGF (15) for a bias voltage  $V \gtrsim \Omega$ . As soon as  $\Gamma, \Delta > V$  one clearly sees the change in sign. On the other hand, at low  $\Gamma, \Delta < V$ , the observed features are nonuniver-

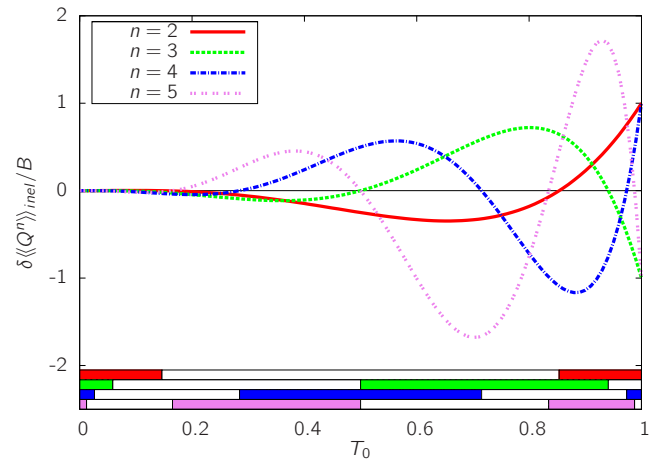


FIG. 2. (Color online) Higher-order cumulants  $\delta\langle Q^n \rangle_{\text{inel}}$  normalized to  $B = g^2 T (V - \Omega) / (2\pi \Gamma^2)$  in the low-energy sector as a function of the bare transmission  $T_0$ . The filled rectangles indicate ranges in which the respective cumulant is positive. The  $n$ th order cumulant has  $n$  zeros in the interval  $T_0 \in [0, 1]$ . The sign changes are spaced symmetrically around  $T_0 = 1/2$ .

sal. Since the sign change occurs only for  $V > \Omega$  and the elastic part is featureless around these critical values of  $\Delta$  and  $\Gamma$ , we believe that the vanishing of the noise correction should be observable in experiments.

This behavior proliferates to cumulants of higher orders. In the  $n$ th order the turning point condition is given by the zeros of an  $n$ th order polynomial. The corresponding solutions for the transmission coefficient are distributed symmetrically around  $T_0 = 1/2$ . This is demonstrated in Fig. 2. One also notes that the cumulants increase drastically toward  $T_0 \rightarrow 1$ . Hence, these effects will be observable in junctions of the type used in Refs. 5–7, where large transmissions  $T_0 \approx 1$  have been observed.

To summarize, we have calculated the charge-transfer statistics of a molecular quantum dot with a single fermionic state coupled to a local Holstein phonon by means of a per-

turbative expansion in the electron-phonon coupling. We found that the FCS is the sum of an adiabatic (mean-field-like) term, an elastic part, and an inelastic term. The latter appears as soon as the applied voltage exceeds the phonon frequency. We found that this inelastic term leaves a characteristic imprint on the noise power as well as on all higher-order cumulants. We expect these features to become observable in experiments in the nearest future.

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