Effect of Electron-Phonon Scattering on Shot Noise in Nanoscale Junctions

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We investigate the effect of electron-phonon inelastic scattering on shot noise in nanoscale junctions in the regime of quasiballistic transport. We predict that when the local thermal energy of the junction is larger than its lowest vibrational mode energy eV_c, the inelastic contribution to shot noise (conductance) increases (decreases) with bias as V ($\sqrt{V}$). The corresponding Fano factor thus increases as $\sqrt{V}$. We also show that the inelastic contribution to the Fano factor saturates with increasing thermal current exchanged between the junction and the bulk electrodes to a value which, for $V \gg V_c$, is independent of bias. These predictions can be readily tested experimentally.

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It is an established fact that for systems with dimensions much longer than the inelastic mean free path λ_{ph} (e.g., a macroscopic sample) steady-state zero-temperature current fluctuations (shot noise) are suppressed by electron-phonon scattering [1,2]. Similarly, for metallic diffusive wires with length much smaller than λ_{ph} (and smaller than the electron-electron scattering length), the Fano factor (i.e., the ratio between shot noise and its Poisson value, $2eI$, where e is the electron charge and I is the current of the system) equals $1/3$ and is not affected by inelastic processes [3]. Systems of nanoscale dimensions may not fall in either one of the above cases. In this instance each electron, on average, releases only a small fraction of its energy to the underlying atomic structure during the time it spends in the junction, making transport quasiballistic [4–10]. However, the current density and, consequently, the power per atom are much larger in the junction compared to the bulk. This leads to heating and inelastic features in the differential conduction which are indeed observed in experiments with metallic quantum point contacts [11–14] and molecular structures [7,9,15–17] as a direct consequence of the interplay between electron and phonon statistics [18]. For these systems it is therefore not obvious what the effect of inelastic scattering on shot noise is.

In this Letter we show analytically that shot noise in quasiballistic nanoscale junctions is enhanced by inelastic scattering whenever electrons have enough energy to excite the phonon modes of the junction. The current instead decreases. As a consequence, the Fano factor increases. We find it increases with bias as $\sqrt{V}$ when the local temperature of the junction is larger than its lowest vibrational mode temperature eV_c/k_B. We also show that with increasing thermal current carried away from the junction to the bulk electrodes, the inelastic contribution to the Fano factor converges to a minimum value independent of bias for $V \gg V_c$. Measurements of the Fano factor and conductance may thus provide information on local temperatures and heat transport mechanisms in these systems. Transport in a model atomic gold point contact will be used to illustrate these findings.

Since the dimensions of the junction are much smaller than λ_{ph} [and the observed inelastic features in quasiballistic systems are very small [11,15,16]] first-order perturbation theory in the electron-phonon coupling captures the dominant contribution to inelastic scattering [19]. This is the contribution we calculate in this Letter.

Let us assume that the junction is connected to two biased bulk electrodes. The electronic states of the full system are thus described by the field operator $\hat{\Psi} = \sum_{E,\alpha=L,R} a_E^{\alpha} \Psi_E^{\alpha}(\mathbf{r}, \mathbf{K}_{\parallel})$, constructed from the single-particle wave functions $\Psi_E^{L(R)}(\mathbf{r}, \mathbf{K}_{\parallel})$ and annihilation operators $a_E^{L(R)}$ corresponding to electrons propagating from the left (right) electrode at energy E. $\mathbf{K}_{\parallel}$ is the component of the momentum parallel to the electrode surface [20]. We also assume that the electrons rapidly thermalize into the bulk electrodes so that their statistics are given by the equilibrium Fermi-Dirac distribution, $f_E^{L(R)} = 1/\{\exp[(E - E_{FL(R)})/k_B T_e] + 1\}$ in the left (right) electrodes with local chemical potential $E_{FL(R)}$, where T_e is the electronic temperature. In the following we will assume that $T_e = 0$ K [21], and the left electrode is positively biased so that $E_{FL} < E_{FR}$. The stationary scattering states $\Psi_E^{L(R)}(\mathbf{r}, \mathbf{K}_{\parallel})$ are eigenstates of an effective single-particle Hamiltonian H_e which may be computed, e.g., using a scattering approach within the static density-functional theory of many-electron systems [20]. The combined dynamics of electrons and phonons is described by the Hamiltonian (atomic units will be used throughout this Letter) [7]

$$H = H_e + H_{\text{ph}} + H_{e\text{-ph}}, \tag{1}$$

where $H_{\text{ph}} = \frac{1}{2}\sum_{i,\mu\in vib} \dot{q}_{i\mu}^2 + \frac{1}{2}\sum_{i,\mu\in vib} \omega_{i\mu}^2 q_{i\mu}^2$ is the phonon contribution, with $q_{i\mu}$ the normal coordinate and $\omega_{i\mu}$ the normal frequency of the vibration labeled by the μth component of the ith ion. $H_{e\text{-ph}}$ describes the electron-phonon interaction and has the following form [7]

$$H_{e\text{-ph}} = \sum_{\alpha,\beta}\sum_{E_1,E_2}\sum_{i\mu,j\nu\in vib}\sqrt{\frac{1}{2\omega_{j\nu}}}A_{i\mu,j\nu}J^{i\mu,\alpha\beta}_{E_1,E_2}a^{\alpha\dagger}_{E_1}a^{\beta}_{E_2} \times (b_{j\nu} + b^{\dagger}_{j\nu}), \tag{2}$$

where $\alpha = L, R$ and $b_{j\nu}$ is the phonon annihilation operator. $\{A_{i\mu,j\nu}\}$ is the transformation matrix that relates Cartesian coordinates to normal coordinates, and $J^{i\mu,\alpha\beta}_{E_1,E_2}$ is the electron-phonon coupling constant which can be directly calculated from the scattering wave functions

$$J^{i\mu,\alpha\beta}_{E_1,E_2} = \int d\mathbf{r}\int d\mathbf{K}_{\parallel}\Psi^{\alpha*}_{E_1}(\mathbf{r},\mathbf{K}_{\parallel})\partial_{\mu}V^{ps}(\mathbf{r},\mathbf{R}_i)\Psi^{\beta}_{E_2}(\mathbf{r},\mathbf{K}_{\parallel}), \tag{3}$$

where we have chosen to describe the electron-ion interaction with pseudopotentials $V^{ps}(\mathbf{r}, \mathbf{R}_i)$ for each ith ion [20].

We use as unperturbed states of the full system (electron plus phonon) the states $|\Psi^{L(R)}_E; n_{j\nu}\rangle = |\Psi^{L(R)}_E(\mathbf{r}, \mathbf{K}_{\parallel})\rangle \otimes |n_{j\nu}\rangle$, where $n_{j\nu}$ is the occupation number of the $j\nu$th normal mode. The first-order perturbation to the wave functions is thus

$$|\Phi^{L(R)}_E; n_{j\nu}\rangle = |\Psi^{L(R)}_E; n_{j\nu}\rangle + |\delta\Psi^{L(R)}_E; n_{j\nu}\rangle, \tag{4}$$

where the first-order correction term is

$$|\delta\Psi^{\alpha}_E; n_{j\nu}\rangle = \lim_{\epsilon\to 0^+}\sum_{\alpha'=L,R}\sum_{m_{j'\nu'}}\int dE' D^{\alpha'}_{E'}\frac{\langle\Psi^{\alpha'}_{E'}; m_{j'\nu'}|H_{\text{el-vib}}|\Psi^{\alpha}_E; n_{j\nu}\rangle|\Psi^{\alpha'}_{E'}; m_{j'\nu'}\rangle}{\varepsilon(E, n_{j\nu}) - \varepsilon(E', m_{j'\nu'}) - i\epsilon}, \tag{5}$$

with $D^{R(L)}_E$ the partial density of states of left (right) moving electrons, and $\varepsilon(E, n_{j\nu}) = E + (n_{j\nu} + 1/2)\omega_{j\nu}$ the energy of state $|\Psi^{\alpha}_E; n_{j\nu}\rangle$. By applying (i) $\langle n_{j\nu} - 1|b_{j\nu}|n_{j\nu}\rangle = \sqrt{n_{j\nu}}$, and $\langle n_{j\nu} + 1|b^{\dagger}_{j\nu}|n_{j\nu}\rangle = \sqrt{1 + n_{j\nu}}$; (ii) $a^{\alpha\dagger}_{E_1}|n^{\beta}_E\rangle = \sqrt{f^{\alpha}_E}|n^{\beta}_E - 1\rangle\delta_{EE_1}\delta_{\alpha\beta}$, and $a^{\alpha}_{E_1}|n^{\beta}_E\rangle = \sqrt{1 - f^{\alpha}_E}|n^{\beta}_E + 1\rangle\delta_{EE_1}\delta_{\alpha\beta}$, we observe that the nonvanishing matrix elements $\langle\Psi^{\alpha'}_{E'}; m_{j'\nu'}|H_{\text{el-vib}}|\Psi^{\alpha}_E; n_{j\nu}\rangle$ correspond to the scattering processes shown in Fig. 1. Carrying out explicitly the integrals in Eq. (5), the corrections to the wave function can be written as

$$|\delta\Psi^{\alpha}_E; n_{j\nu}\rangle = (B^{\alpha}_{j\nu,1} + B^{\alpha}_{j\nu,3})|\Psi^{\alpha}_{E+\omega_{j\nu}}; n_{j\nu} + 1\rangle + (B^{\alpha}_{j\nu,2} + B^{\alpha}_{j\nu,4})|\Psi^{\alpha}_{E-\omega_{j\nu}}; n_{j\nu} - 1\rangle, \tag{6}$$

where $B^{\alpha}_{j\nu,1}$, $B^{\alpha}_{j\nu,2}$, $B^{\alpha}_{j\nu,3}$, and $B^{\alpha}_{j\nu,4}$ correspond to the diagrams depicted in Fig. 1. For $|\delta\Psi^R_E; n_{j\nu}\rangle$, the coefficients are given by:

$$B^R_{j\nu,1(2)} = i\pi\sum_{i\mu}\sqrt{\frac{1}{2\omega_{j\nu}}}A_{i\mu,j\nu}J^{i\mu,LR}_{E\pm\omega_{j\nu},E}D^L_{E\pm\omega_{j\nu}}\sqrt{(\delta + n_{j\nu})f^R_E(1 - f^L_{E\pm\omega_{j\nu}})}, \tag{7}$$

and

$$B^R_{j\nu,3(4)} = -i\pi\sum_{i\mu}\sqrt{\frac{1}{2\omega_{j\nu}}}A_{i\mu,j\nu}J^{i\mu,RL}_{E\pm\omega_{j\nu},E}D^L_{E\pm\omega_{j\nu}}\sqrt{(\delta + n_{j\nu})f^L_E(1 - f^R_{E\pm\omega_{j\nu}})}, \tag{8}$$

where $\delta = 1$ and "$-$" sign are for the scattering diagrams (a) and (c); $\delta = 0$ and "$+$" sign for diagrams (b) and (d). Similarly, the coefficients in $|\delta\Psi^L_E; n_{j\nu}\rangle$ have the forms $B^L_{j\nu,k} = B^R_{j\nu,k}(L\leftrightharpoons R)$, where $k = 1, \ldots, 4$; the notation $(L\leftrightharpoons R)$ means interchange of labels R and L.

At $T_e = 0$ K the first-order correction to the current is thus:

$$I = -i\int_{E_{FL}}^{E_{FR}} dE\int d\mathbf{R}\int d\mathbf{K}_{\parallel}\tilde{I}^{RR}_{E,E}\left[1 - \sum_{j\nu}(\langle|B^R_{j\nu,1}|^2\rangle + \langle|B^R_{j\nu,2}|^2\rangle)\right], \tag{9}$$

where $\tilde{I}^{\alpha\beta}_{E,E} \equiv (\Psi^{\alpha}_E)^*\partial_z(\Psi^{\beta}_E) - \partial_z(\Psi^{\alpha}_E)^*(\Psi^{\beta}_E)$ and $\langle\rangle$ indicates the ensemble average over phonon states. Here we assume that the ions of the junction reach thermal equilibrium with a well-defined local temperature T_w such that ensemble averages of phonon states are $\langle n_{j\nu}\rangle = 1/[\exp(\omega_{j\nu}/k_BT_w) - 1]$ and $\langle n^2_{j\nu}\rangle = [\exp(\omega_{j\nu}/k_BT_w) + 1]/[\exp(\omega_{j\nu}/k_BT_w) - 1]^2$ [7,9]. Equation (9) has been simplified by using (i) $\tilde{I}^{RR}_{E\pm\omega_{j\nu},E\pm\omega_{j\nu}} \simeq \tilde{I}^{RR}_{E,E}$, valid for energies close to the chemical potentials; and (ii) $\tilde{I}^{RR}_{E,E} = -\tilde{I}^{LL}_{E,E}$, a direct consequence of time-reversal symmetry. The current is therefore reduced by inelastic effects.

Let us now calculate the corresponding correction to shot noise. We have previously shown that shot noise can be written in terms of single-particle scattering states as [22,23]

$$S = \int_{E_{FL}}^{E_{FR}} dE\left|\int d\mathbf{R}\int d\mathbf{K}\tilde{I}^{LR}_{E,E}\right|^2, \tag{10}$$

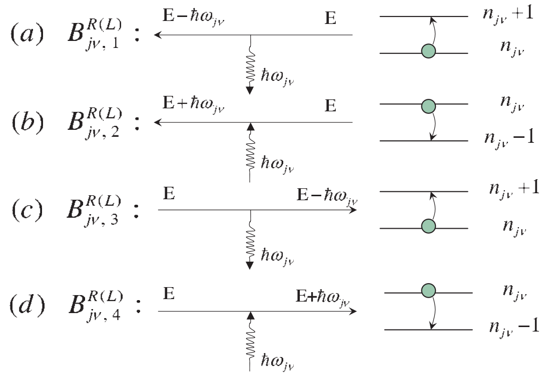

FIG. 1 (color online). Feynman diagrams and corresponding amplitudes (see text) of the main electron-phonon scattering mechanisms contributing to the correction of the current and noise.

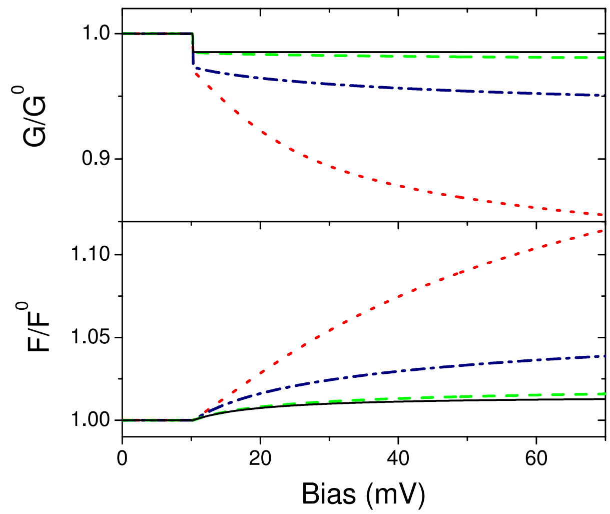

FIG. 2 (color online). Top panel: ratio of the total conductance G of an atomic gold point contact and its value in the absence of inelastic effects G^0 as a function of bias for different values of thermal current coefficient (see text): $A_{\rm th} = 10^{-19}$ (dotted line), 10^{-17} (dot-dashed line), 10^{-15} (dashed line), and ∞ (solid line) dyn/(sK^4). Bottom panel: corresponding Fano factor ratio.

which reduces to the well-known formula $S \propto \sum_i T_i(1 - T_i)$ when the eigenchannels transmission probabilities T_i are extracted from the single-particle states with independent transverse momenta [1,22,23]. Replacing (4) into (10) we get

$$S = \int_{E_{FL}}^{E_{FR}} dE \left| \int d\mathbf{R} \int d\mathbf{K} \tilde{I}^{LR}_{E,E} \right|^2 \times \left[1 + \sum_{j\nu;k=1,2} (\langle |B^R_{j\nu,k} B^{L*}_{j\nu,k}|^2 \rangle) \right]. \tag{11}$$

Since the summation over vibrational modes contains only positive terms, shot noise is *enhanced* by electron-phonon inelastic effects in the quasiballistic regime. Therefore, the Fano factor F normalized to the corresponding value in the absence of electron-phonon interactions (F^0) is

$$F/F^0 = \frac{\int_{E_{FL}}^{E_{FR}} dE [1 + \sum_{j\nu,k=1,2} (\langle |B^R_{j\nu,k} B^{L*}_{j\nu,k}|^2 \rangle)]}{\int_{E_{FL}}^{E_{FR}} dE [1 - \sum_{j\nu,k=1,2} \langle |B^R_{j\nu,k}|^2 \rangle]}, \tag{12}$$

which *increases* with electron-phonon scattering.

Note that due to the orthogonality of phonon states, the absolute value of the correction to shot noise is smaller than that to the current [cf. Eqs. (9) and (11)]. Note also that conservation of energy and the Pauli exclusion principle play an important role. The former dictates an onset bias V_c for inelastic contributions; the latter prohibits the scattering processes depicted in Figs. 1(c) and 1(d) at $T_e = 0$ K.

These results are illustrated in Fig. 2 where the inelastic contribution to the conductance and Fano factor are plotted for a gold atom placed in the middle of two bulk gold electrodes (represented with ideal metals, jellium model, $r_s \approx 3$). Details of the calculations can be found in Refs. [7,20]. In the absence of electron-phonon interactions, the unperturbed differential conductance G^0 is about 1.1 (in units of $2e^2/h$) and the Fano factor is $F^0 \simeq 0.14$ [22] in the bias range of Fig. 2. Inelastic effects cause a discontinuity in the conductance, and a variation of the Fano factor ratio [Eq. (12)], at a bias $V_c \approx 11$ mV, corresponding to the energy of the lowest longitudinal mode of the system. In addition, the above inelastic corrections depend on the local temperature of the junction T_w [see Eqs. (7) and (8)] which, in turn, is the result of the competition between the rate of heat generated locally in the nanostructure and the thermal current $I_{\rm th}$ carried away into the bulk electrodes [4–7,9,10]. The latter has a temperature dependence of $I_{\rm th} = A_{\rm th} T_w^4$ [24], where the constant $A_{\rm th}$ depends on the details of the coupling between the local modes of the junction and the modes of the bulk electrodes. At steady state this thermal current has to balance the power generated in the nanostructure, which is a small fraction of the total power of the circuit $\frac{V^2}{R}$ (V is the bias, R is the resistance) [4,7].

The larger $A_{\rm th}$, the larger the heat dissipated into the bulk and, thus, the lower the local temperature T_w [25]. In the limit of infinite $A_{\rm th}$, i.e., $T_w = 0$, at any given bias larger than V_c, electrons can only emit phonons [$n_{j\nu} = 0$ in Eqs. (7) and (8)]. The inelastic contribution to the conductance and Fano factor, therefore, saturate to a specific value (see Fig. 2). We can derive both the bias dependence and this saturation value, to first order in the bias, as follows.

By equating the thermal current $I_{\rm th}$ to the power generated in the junction, it is easy to show that $T_w = \alpha\sqrt{V}$ [6,26], where the constant α depends on the details of the thermal contacts between the junction and electrodes. Let us assume for simplicity a single phonon mode of frequency ω. For $T_w > \omega/k_B$, we expand $\langle n_{j\nu}\rangle \approx k_B T_w/\omega$ in Eq. (9). We then get

$$\frac{G}{G^0} \simeq 1 - \alpha \frac{3}{2}\frac{k_B}{\omega}\gamma_I \theta(V - V_c)\sqrt{V}, \qquad (13)$$

where $\theta(V - V_c)$ is the Heaviside function; $\gamma_I = |[d(I - I^0)/dV]/(dI^0/dV)|$ is the relative change in conductance due to inelastic effects at V_c [its value is about 1% for the specific case, in agreement with experiments on similar systems [7,11]]. The inelastic contribution to the conductance thus decreases with bias as $\sqrt{V}$. This square-root dependence is clear in Fig. 2 for $A_{\rm th} < 10^{-15}$ dyn/(sK^4) which corresponds to temperatures for which the condition $T_w > \omega/k_B$ is satisfied.

The same analysis can be applied to shot noise. In Eq. (11), for $T_w > \omega/k_B$ we expand $\langle n_{j\nu}^2\rangle \approx 2(k_B T_w/\omega)^2$ which leads to

$$\frac{S}{S^0} \simeq 1 + 2\alpha^2\left(\frac{k_B}{\omega}\right)^2 \gamma_s \theta(V - V_c)(V - V_c), \qquad (14)$$

where $\gamma_S = |[d(S - S^0)/dV]/(dS^0/dV)|$ is the relative change of shot noise due to inelastic effects at $V = V_c$ (it is about 0.04% for the specific gold junction). The inelastic correction to shot noise thus increases linearly with bias for $T_w > \omega/k_B$. Consequently, $F/F^0 \propto \sqrt{V}$ as it is also evident from Fig. 2.

In the opposite limit of perfect heat dissipation in the bulk electrodes, i.e., for $T_w \to 0$ [see Fig. 2, $A_{\rm th} \to \infty$ dyn/(sK^4)], then from Eqs. (7) and (8) it is easy to prove that $I/I_0 = 1 - \theta(V - V_c)\gamma_I(V - V_c)/V$ and $S/S_0 = 1 + \gamma_S[(V - V_c)/V]\theta(V - V_c)$. Therefore,

$$F/F^0 = \frac{1 + \gamma_S[(V - V_c)/V]\theta(V - V_c)}{1 - \gamma_I[(V - V_c)/V]\theta(V - V_c)}, \qquad (15)$$

which tends to the constant value $F/F^0 \to (1 + \gamma_S)/(1 - \gamma_I)$ as $V \gg V_c$. The predictions reported in this Letter should be readily tested experimentally.

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