Phonon-Assisted Current Noise in Molecular Junctions

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We investigate the effects of phonon scattering on the electronic current noise through nanojunctions using the nonequilibrium Green's functions formalism extended to include the counting field. In the case of weak electron-phonon coupling and a single broad electronic level, we derive an analytic expression for the current noise at arbitrary temperature and identify physically distinct contributions based on their voltage dependence. We apply our theory to the experimentally relevant case of a D_2 molecule placed in a break junction and predict a significant inelastic contribution to the current noise.

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Introduction.—The fabrication of atomically sharp contacts has opened up the possibility of creating junctions formed by a single molecule bridging metal electrodes [1]. However, the inherent complexity of this emerging field poses fundamental challenges and makes junctions formed by very simple molecules (e.g., hydrogen [2–4] or water [5]) an invaluable test bed from both the experimental and the theoretical point of view. In these systems, inelastic effects due to the interaction between transport electrons and molecular vibrational modes (phonons) result in an abrupt change of the differential conductance at the onset of the phonon emission. These features have been exploited to establish unambiguously the presence of the molecule in the contact [2] and, when combined with shot-noise measurements [4,6], they allow for a detailed characterization of the junction.

Theoretical descriptions of inelastic transport through nanojunctions have so far focused mainly on the current-voltage characteristics, while less attention has been paid to the study of noise. Phonon-scattering effects on the differential conductance have been addressed both with *ab initio* methods [7–12] and with simplified (one-level) models [13–16]. Noise calculations based on one-level models have also been put forward within the rate equation approach [13,14] or within the nonequilibrium Green's functions (NGF) formalism [17] with a mean-field-like approximation for the noise. In this work, we study inelastic effects on the current noise with the NGF approach taking consistently into account all the correlations due to phonon-assisted scattering up to a given order in the e-ph interaction. We apply our theory to the case of a hydrogen-bridge junction and predict a significant inelastic contribution to the current noise.

Model and methods.—The system we consider can be represented as a central device region which is tunnel-coupled to noninteracting metallic leads $\hat{H} = \hat{H}_C + \hat{H}_{L,R} + \hat{H}_T$. Neglecting the spin degree of freedom, the central region can be described by the Hamiltonian $\hat{H}_C = \hat{H}_0 + \sum_\ell \hbar\omega_\ell \hat{b}_\ell^\dagger \hat{b}_\ell + \sum_\ell \sum_{i,j} M_\ell^{ij} \hat{d}_i^\dagger \hat{d}_j (\hat{b}_\ell^\dagger + \hat{b}_\ell)$, where $\hat{d}_i^\dagger$ and $\hat{b}_\ell^\dagger$ are the electron and phonon creation operators, $\hat{H}_0 = \sum_{i,j} H_0^{ij} \hat{d}_i^\dagger \hat{d}_j$ is the single-particle effective Hamiltonian of the electrons moving in a static arrangement of atomic nuclei, and $\mathbf{M}_\ell$ is the e-ph coupling matrix for the ℓ-th phonon mode. Here, boldface notation stands for matrix over electronic space. The leads and tunneling Hamiltonians are given by $\hat{H}_{L,R} = \sum_{k,\alpha=L,R} \varepsilon_{\alpha,k} \hat{c}_{\alpha,k}^\dagger \hat{c}_{\alpha,k}$ and $\hat{H}_T = \sum_{k,\alpha=L,R} (V_{\alpha,k}^i \hat{c}_{\alpha,k}^\dagger \hat{d}_i + \text{H.c.})$. The states in the leads are occupied according to the Fermi distribution $f_\alpha(\varepsilon) = [1 + e^{\beta(\varepsilon - \mu_\alpha)}]^{-1}$, where $\beta = 1/k_B T$ is the inverse temperature and μ_α is lead-α chemical potential, with $\mu_L - \mu_R = \text{eV}$ fixed by the applied bias voltage.

To calculate the current and the noise, we employ the extended NGF technique [18] to find the cumulant generating function $\mathcal{S}(\lambda) = I/(ie)\lambda + S/(2e^2)\lambda^2 + \cdots$, from which the current I and the noise S can be calculated straightforwardly. The key idea [19] is to modify the Hamiltonian by adding a time-dependent phase $\lambda(t)/2$ to the tunneling matrix elements $V_{L,k}^i$, with $\lambda(t) = \pm\lambda$ for t on the upper and the lower Keldysh contour, respectively. It has been shown in Ref. [20] for the Anderson model that $\partial_\lambda \mathcal{S}(\lambda)$ is related to the Keldysh Green's function $G_\lambda(t, t') = -i\langle \mathcal{T}_C \hat{d}(t) \hat{d}^\dagger(t') \rangle_\lambda$, where the expectation value is now evaluated in the presence of $\lambda(t)$. Generalizing that result to the multilevel case, we write

$$\frac{\partial \mathcal{S}(\lambda)}{\partial \lambda} = \int \frac{d\varepsilon}{2\pi\hbar} \text{Tr}\{\mathbf{\Gamma}_L [e^{-i\lambda}(1 - f_L)\mathbf{G}_\lambda^{-+} + e^{i\lambda} f_L \mathbf{G}_\lambda^{+-}]\}. \quad (1)$$

Here, $\Gamma_\alpha^{ij}(\varepsilon) = 2\pi \sum_k V_{\alpha,k}^i V_{\alpha,k}^{j^*} \delta(\varepsilon - \varepsilon_{k,\alpha})$ is the level broadening due to the coupling to lead-α and $\mathbf{G}_\lambda^{-+}$, $\mathbf{G}_\lambda^{+-}$ are appropriate components of the solution of the nonequilibrium Dyson equation $\check{\mathbf{G}}_\lambda = \check{\mathbf{g}}_\lambda + \check{\mathbf{g}}_\lambda \check{\mathbf{\Sigma}}_\lambda \check{\mathbf{G}}_\lambda$. The check sign indicates matrices in the Keldysh space, and the superscripts − (+) correspond to the forward (backward) branch of the Keldysh contour. The matrix $\check{\mathbf{g}}_\lambda$ is Green's function of the system in the presence of the leads and of the counting field but without the e-ph interaction. Its inverse is given by

$$\check{\mathbf{g}}_\lambda^{-1}(\varepsilon) = \begin{pmatrix} \varepsilon\mathbf{1} - \mathbf{H}_0 - i\sum_\alpha \mathbf{\Gamma}_\alpha[f_\alpha(\varepsilon) - 1/2] & i\mathbf{\Gamma}_L e^{i\lambda} f_L(\varepsilon) + i\mathbf{\Gamma}_R f_R(\varepsilon) \\ -i\mathbf{\Gamma}_L e^{-i\lambda}[1 - f_L(\varepsilon)] - i\mathbf{\Gamma}_R[1 - f_R(\varepsilon)] & -\varepsilon\mathbf{1} + \mathbf{H}_0 - i\sum_\alpha \mathbf{\Gamma}_\alpha[f_\alpha(\varepsilon) - 1/2] \end{pmatrix}. \quad (2)$$

As $\check{\mathbf{g}}_\lambda$ already includes the coupling to the leads, $\check{\mathbf{\Sigma}}_\lambda$ is the self-energy solely due to the e-ph coupling. Being interested in the weak coupling limit, we expand the Dyson equation to the lowest (second) order in the e-ph coupling $\check{\mathbf{G}}_\lambda \approx \check{\mathbf{g}}_\lambda + \check{\mathbf{g}}_\lambda \check{\mathbf{\Sigma}}_\lambda^{(2)} \check{\mathbf{g}}_\lambda$, where $\mathbf{\Sigma}_\lambda^{(2)}$ is given by the Fock diagram (η, $\bar{\eta} = \pm$)

$$\mathbf{\Sigma}_\lambda^{(2)\eta\bar{\eta}}(\varepsilon) = i\sum_\ell \int \frac{d\varepsilon'}{2\pi} d_\ell^{\eta\bar{\eta}}(\varepsilon - \varepsilon')\mathbf{M}_\ell \mathbf{g}_\lambda^{\eta\bar{\eta}}(\varepsilon')\mathbf{M}_\ell. \quad (3)$$

The Hartree term has been neglected since it cannot contribute by any truly dynamical features in which we are primarily interested. Above, $d_\ell^{\eta\bar{\eta}}(\varepsilon)$ stand for *free* phonon Green's functions for the ℓ-th phonon mode $d_\ell^{\pm\pm}(\varepsilon) = \sum_{s=\pm}[-i\pi(2\mathcal{N}_\ell + 1)\delta(\varepsilon + s\hbar\omega_\ell) \pm \mathcal{P}\frac{s}{\varepsilon + s\hbar\omega_\ell}]$ and $d_\ell^{+,\pm}(\varepsilon) = -2\pi i[(\mathcal{N}_\ell + 1)\delta(\varepsilon \pm \hbar\omega_\ell) + \mathcal{N}_\ell\delta(\varepsilon \mp \hbar\omega_\ell)]$, with $\mathcal{N}_\ell$ the (generally nonequilibrium) occupation of mode ℓ.

Truncating the Dyson equation to the second order in $\mathbf{M}_\ell$ directly yields the expressions for $\check{\mathbf{G}}_{\lambda=0}$ and $\partial_\lambda \check{\mathbf{G}}_\lambda|_{\lambda=0}$, which are the ingredients to evaluate the current and the noise. Integration over energy can be performed analytically assuming the electronic structure to be slowly changing over few multiples of a typical phonon energy around the Fermi level E_F and approximating $\mathbf{\Gamma}_\alpha(\varepsilon) \approx \mathbf{\Gamma}_\alpha(E_F)$ and $\mathbf{g}_{\lambda=0}^r(\varepsilon) \approx \mathbf{g}_{\lambda=0}^r(E_F)$ [8–11].

Physically important nonequilibrium phonon heating effects [21] can be qualitatively taken into account by a rate equation for the average phonon occupation number $\mathcal{N}_\ell$ [7], which can be viewed as a kinetic-equation-like approximation to the full NGF studies [12,15]. In the broad-level approximation introduced above, this leads to a bias-dependent occupation number $\mathcal{N}_\ell(V) = n_B(\omega_\ell) + \alpha_d n_\ell(V)$, where $n_B(\omega) = 1/(e^{\beta\hbar\omega} - 1)$ is the Bose distribution, α_d is a parameter which depends on strength of some external phonon damping, and $n_\ell(V)$ takes into account the power dissipated by the transport electrons into the phonon mode [8]. In the following, we will focus on the two opposite regimes of (i) thermally equilibrated phonons ($\alpha_d = 0$), and (ii) nonequilibrated phonons ($\alpha_d = 1$). In this case, it is $n_\ell(V) \approx (|eV|/\hbar\omega_\ell - 1)\theta(|eV| - \hbar\omega_\ell)/4$ for $k_B T \ll \hbar\omega_\ell$ [11].

Analysis.—For sake of clarity, we focus here only on the case of a single electronic level ε_0 with symmetric coupling to both leads $\Gamma_L = \Gamma_R = \Gamma$ and coupled to a single phonon mode with frequency ω_0, occupation $\mathcal{N}_0$, and coupling constant M. Already such a simple model reveals many essential features of inelastic transport through nanojunctions such as the phonon-induced step behavior of the differential conductance [5,16]. Before focusing on the inelastic corrections to the noise, we shortly reexamine the results for the current. We find that the current through the device is given by $I = I_{\rm el} + I_{\rm inel}$, with $I_{\rm el} = (e^2/h)\mathcal{T}V$ and

$$I_{\rm inel} = \frac{e\gamma_{\rm eph}\omega_0}{2\pi}\Big[(1 - 2\mathcal{T})\frac{W(\bar{V} - 1) - W(\bar{V} + 1)}{2} + (2\mathcal{N}_0 + 1)(3 - 4\mathcal{T})\bar{V}\Big], \quad (4)$$

with the reduced voltage $\bar{V} = {\rm eV}/\hbar\omega_0$, the dimensionless e-ph coupling $\gamma_{\rm eph} = M^2\mathcal{T}^2/\Gamma^2$, and $W(x) = x\coth(\beta\hbar\omega_0 x/2)$. Here, $\mathcal{T}$ is the elastic transmission coefficient $\mathcal{T} = |G^r|^2\Gamma^2 = \Gamma^2/(\Delta^2 + \Gamma^2)$ and $\Delta = (E_F - \varepsilon_0)$ gives the position of the single level with respect to the Fermi energy. The inelastic current results then from the sum of two contributions with distinct behavior with respect to the bias voltage: while the first term of Eq. (4), which is responsible for the step features in the nonlinear conductance, saturates to constant values for $|\bar{V}| > 1$, the second one grows linearly with V for $\mathcal{N}_0 = n_B(\omega_0)$ and quadratically in the case of nonequilibrated phonons. This second term has a clear physical interpretation in terms of electrons experiencing the coupling to the phonon as a stochastic quasistatic shift in the energy of the level. This in turn affects the transmission coefficient, which becomes dependent on the displacement of the oscillator $\mathcal{T}(Q)$. Averaging over Q and retaining only terms to the second order in M, one obtains

$$\langle\mathcal{T}(Q)\rangle = \Big\langle\frac{\Gamma^2}{(\Delta - MQ)^2 + \Gamma^2}\Big\rangle \approx \mathcal{T} + \gamma_{\rm eph}(3 - 4\mathcal{T})\langle Q^2\rangle, \quad (5)$$

where $\langle\cdot\rangle$ indicates the average over a stationary distribution of the oscillator so that $\langle Q\rangle = 0$ and $\langle Q^2\rangle = (2\mathcal{N}_0 + 1)$. We can therefore interpret the last term of Eq. (4) in terms of an elasticlike contribution with averaged transmission over the fluctuating position of the oscillator [22].

We now turn to the main result of our work which is the phonon-assisted noise. The current noise through the device is given by $S = S_{\rm el} + S_{\rm inel}$, with the standard expression for the elastic noise $S_{\rm el} = (e^2/h)\{\mathcal{T}^2/\beta + \mathcal{T}(1 - \mathcal{T})\hbar\omega_0 W(\bar{V})\}$ [23] and with

$$S_{\rm inel} = \frac{e^2\gamma_{\rm eph}\omega_0}{2\pi}\Big\{\Big[\frac{c_0}{\beta\hbar\omega_0} + c_1 W(\bar{V})\Big] + \kappa_0 - \kappa_1\Big[\sum_{s=\pm 1}\frac{W(\bar{V} + s)}{2} - W(\bar{V})\Big] - \frac{\kappa_2}{\beta\hbar\omega_0}\sum_{s=\pm 1} sW'(\bar{V} + s)\Big\} \quad (6)$$

being the correction due to inelastic scattering. Here, $W'(x) = dW/dx$ and the coefficients read $c_0 = 4(2\mathcal{N}_0 + 1)\mathcal{T}(5 - 6\mathcal{T})$, $c_1 = (2\mathcal{N}_0 + 1)(12\mathcal{T}^2 - 14\mathcal{T} + 3)$, $\kappa_0 = 4\mathcal{T}(1 - \mathcal{T})(1 + 2\mathcal{N}_0)\{[1 + 2n_B(\omega_0)] - 2/\beta\hbar\omega_0\} - (1 - 2\mathcal{T})^2$, $\kappa_1 = 4\mathcal{T}(1 - \mathcal{T})[1 + 2n_B(\omega_0)] - (1 - 2\mathcal{T})^2(1 + 2\mathcal{N}_0)$, and, finally, $\kappa_2 = 2\mathcal{T}(1 - 2\mathcal{T})$. In the zero bias limit, $S_{\rm inel}$ satisfies the fluctuation-dissipation theorem, i.e., $S_{\rm inel}|_{V=0} = 2\mathcal{G}_{\rm inel}/\beta$, where $\mathcal{G}_{\rm inel} = (\partial I_{\rm inel}/\partial V)|_{V=0}$ is the inelastic correction to the linear conductance.

Similarly as for the current, we interpret the first term of Eq. (6) as a quasistatic correction to the elastic noise due to averaged transmission over the displacement of the oscillator. In fact, along the same line which lead to Eq. (5), it is easy to see that $\gamma_{\rm eph}c_0$ and $\gamma_{\rm eph}c_1$ correspond exactly to the contribution of order M^2 to $\langle T(Q)^2\rangle$ and $\langle T(Q)[1 - T(Q)]\rangle$, respectively. The remaining terms are dynamic contributions which take into account the phonon exchange effects. Analogously to the current, the quasistatic contribution has a distinct voltage behavior compared to the dynamic one for large voltage, being the only one which does not saturate for $|\bar{V}| > 1$.

In the limit of zero temperature, Eq. (7) simplifies noticeably becoming

$$S_{\rm inel}|_{T=0} = \frac{e^2\gamma_{\rm eph}\omega_0}{2\pi}\{\kappa_0|_{T=0} + c_1|_{T=0}|\bar{V}| - \kappa_1|_{T=0}(1 - |\bar{V}|)\theta(1 - |\bar{V}|)\}. \qquad (7)$$

The inelastic noise is then a piecewise function characterized by the coefficient $c_1|_{T=0}$ for $|\bar{V}| > 1$ and by the combination $(c_1 + \kappa_1)|_{T=0}$ for $|\bar{V}| < 1$, which takes into account the competition between quasistatic contribution and the dynamic one. The signs of these coefficients determine whether $S_{\rm inel}$ is an increasing or decreasing function of voltage, leading to the rich behavior presented in Fig. 1. Interestingly, depending on the value of $\mathcal{T}$, both $(c_1 + \kappa_1)|_{T=0}$ and $c_1|_{T=0}$ can be negative, resulting in a negative contribution to the inelastic noise. In other words, for a rather wide range of transmissions, phonon-scattering events lead to suppression of the current noise through the device. Note that both c_1 and κ_1 may depend on voltage via $\mathcal{N}_0$. However, since the phonon heating becomes effective only above the phonon emission threshold, $S_{\rm inel}$ is always a linear function for $|\bar{V}| < 1$, while energy accumulation into the phonon mode results in the quadratic increase of $S_{\rm inel}$ for $|\bar{V}| > 1$ in the case of no external damping (nonequilibrated phonons, $\alpha_d = 1$).

For finite temperatures, qualitatively new features appear (see Fig. 2). For small temperatures, the curves get rounded around $\bar{V} = 1$ and for high transmission even an additional dip occurs [see Fig. 2(a)]. The changes become more pronounced if the temperature is of the order of the phonon frequency, when the kinks of $S_{\rm inel}$ are largely washed out. Nevertheless, it is still possible for a wide set of parameters to have a negative inelastic correction to the noise and even a sign change at some finite $|\bar{V}|$ [see Fig. 2(b)].

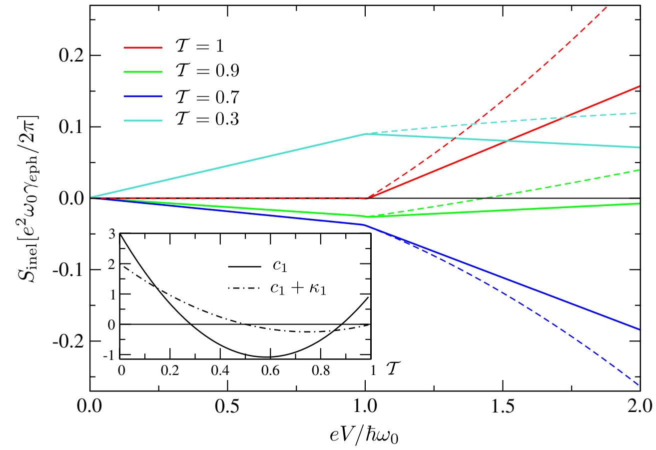

FIG. 1 (color online). Inelastic noise $S_{\rm inel}$ as a function of the bias voltage at zero temperature $T = 0$ for different values of the transmission coefficient. Both cases of equilibrated ($\alpha_d = 0$, thick lines) and nonequilibrated phonons ($\alpha_d = 1$, dashed lines) are shown. Inset: Plot of $c_1|_{\{T=0,|eV|=\hbar\omega_0\}}$ (full line) and $(c_1 + \kappa_1)|_{\{T=0,eV=0\}}$ (dash-dotted line) as a function of $\mathcal{T}$.

Results.—We apply now our formulas to the case of a single hydrogen molecule between platinum atomic contacts [2]. Experimentally, it has been shown that hydrogen can form a stable bridge between Pt electrodes with conductance close to the quantum unit [2,3] carried dominantly by a single, almost transparent channel [4]. Such a picture has been confirmed by first principle calculations showing that a single conductance channel forms due to strong hybridization between the H_2 antibonding state and

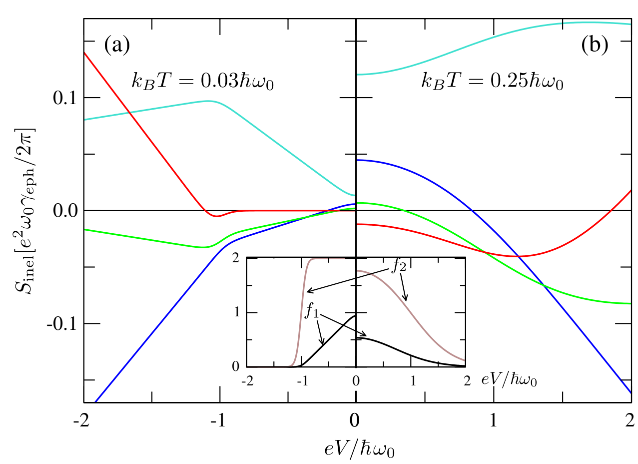

FIG. 2 (color online). Temperature dependence of inelastic noise. (a) $S_{\rm inel}$ for the case of equilibrated phonons ($\alpha_d = 0$) at $k_BT = 0.03\hbar\omega_0$ (a typical experimental value [3,8]). Different lines correspond to various values of the transmission coefficient $\mathcal{T}$ (color code identical to Fig. 1). (b) Same as in (a) but at higher temperature $k_BT = 0.25\hbar\omega_0$. Inset: Plots of $f_1(\bar{V}) = \frac{1}{2}[W(\bar{V} + 1) - 2W(\bar{V}) + W(\bar{V} - 1)]$ and $f_2(\bar{V}) = W'(\bar{V} + 1) - W'(\bar{V} - 1)$ at $k_BT = 0.03\hbar\omega_0$ (left panel) and $k_BT = 0.25\hbar\omega_0$ (right panel). These functions characterize the dynamic contributions to $S_{\rm inel}$ in Eq. (6).

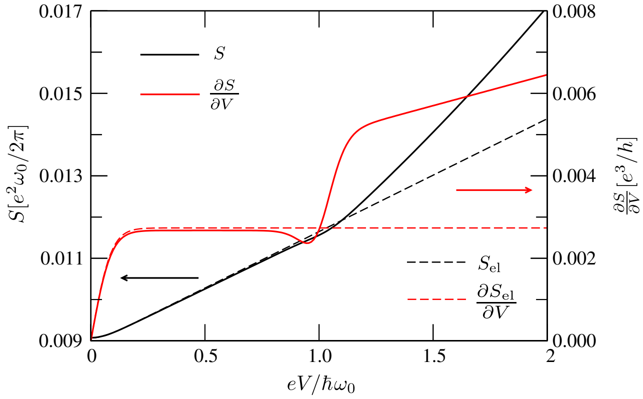

FIG. 3 (color online). Total current noise $S = S_{\rm el} + S_{\rm inel}$ through a D_2 molecule and its derivative $\partial S/\partial V$ as a function of voltage. The elastic contributions $S_{\rm el}$ and $\partial S_{\rm el}/\partial V$ are plotted as dashed lines for comparison. Parameter values $k_B T = 0.029\hbar\omega_0$, $\hbar\omega_0 = 50$ MeV, $\tau = 0.9825$, $\gamma_{eph} = 0.011$, and $\alpha_d = 1$ are taken from Ref. [8] and correspond to the conductance measurements in Ref. [3]. The noise level at $\mathrm{eV} = \hbar\omega_0 = 50$ MeV corresponds to experimentally accessible 3×10^{-27} A^2/Hz. Phonon heating ($\alpha_d = 1$) responsible for the finite slope of $\partial S/\partial V$ for $|\mathrm{eV}| > \hbar\omega_0$ must be included because of the large mass mismatch between the D_2 molecule and Pt atoms.

the Pt metal states, while the bonding state is not involved in the transport [24].

Figure 3 represents our prediction for the phonon-assisted noise through a D_2 junction, where typical values for $\mathcal{T}$, $k_B T$, and γ_{eph} have been taken from Ref. [8] and correspond to the experimental data of Ref. [3]. One main message of our work is that, despite of the very weak e-ph coupling, inelastic corrections give a sizable contribution to the total noise through a D_2 junction for $|\mathrm{eV}| > \hbar\omega_0$. On the other hand, inelastic corrections are negligible for $|\mathrm{eV}| < \hbar\omega_0$, thus justifying the interpretation of noise measurement in this regime in terms of elastic theory (as done in [4]).

In conclusion, we have presented a perturbative scheme for the calculation of the inelastic contribution to the current noise in systems with weak e-ph interaction. In the experimentally relevant case of a single broad level, we have derived an analytic expression for the inelastic noise at arbitrary temperature and distinguished terms that correspond to simple renormalization of the transmission coefficient from those which contain true dynamical effects. Applying our theory to the case of a D_2 junction, we predict a sizable contribution to the total noise due to inelastic processes. Our scheme can be straightforwardly extended beyond the present model to cases with multiple electronic levels and phonon modes, asymmetric coupling to leads, energy-dependent transmission, and/or moderate e-ph coupling with application in current *ab initio* methods [8–11].

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Note added.—After submission of the present Letter, two related works were published [25].

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