Nonequilibrium Charge Susceptibility and Dynamical Conductance: Identification of Scattering Processes in Quantum Transport

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We calculate the nonequilibrium charge transport properties of nanoscale junctions in the steady state and extend the concept of charge susceptibility to the nonequilibrium conditions. We show that the nonequilibrium charge susceptibility is related to the nonlinear dynamical conductance. In spectroscopic terms, both contain the same features versus applied bias when charge fluctuation occurs in the corresponding electronic resonances. However, we show that, while the conductance exhibits features at biases corresponding to inelastic scattering with no charge fluctuations, the nonequilibrium charge susceptibility does not. We suggest that measuring both the nonequilibrium conductance and charge susceptibility in the same experiment will permit us to differentiate between different scattering processes in quantum transport.

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Recent developments in modern techniques of microscopic manipulation and nanotechnologies enable us to build functional nanoscale systems, for example, electronic nanodevices or molecular motors [1–5]. In such systems, classical equilibrium thermodynamics is not suited to describe the quantization of the charge or heat flow. The properties of such devices differ from their equilibrium counterparts since nonequilibrium quantum and nonlinear effects dominate. The concepts of conventional statistical mechanics and linear-response theory for these small systems need to be substituted with those of nonequilibrium quantum statistical mechanics [6–8]. This is the appropriate framework for dealing with nanoscale systems driven out of equilibrium, especially when one wants to design or control these systems as heat engines or electromechanical devices.

Extending the concept of equilibrium statistical mechanics (linear-response theory, response functions, and fluctuation theorems) to the nonequilibrium (NE) conditions has seen a recent growing interest [9–11]. It is known from linear-response theory that there exists some relationship between different response functions, like, for example, the density-density, current-density, or current-current correlation functions at equilibrium [12]. However, there is no reason why these relationships should hold at NE. Motivated by understanding these NE properties and their use in practical nanoscale devices, we focus in this Letter on a specific physical property: the electronic transport. In particular, we consider the relationship between the electrical conductance and the charge susceptibility in nanoscale junctions. We provide a definition for the NE charge susceptibility, which can be measured in experiments, and examine in detail its relationship with the full nonlinear dynamical conductance.

We show that the nonequilibrium charge susceptibility and the dynamical conductance of such a system are related to each other, although in a different manner than at equilibrium. At finite bias, they both contain information about the charge fluctuation (induced by the bias) in the electronic resonances. However, the NE charge susceptibility does not contain information about purely inelastic scattering processes which do not involve charge fluctuations. By measuring both the conductance and the NE charge susceptibility in the same experiment, one can identify the nature of scattering processes involved in transport through nanoscale junctions.

We illustrate this property with numerical calculations for a model of a single-molecule nanojunction in the presence of electron-phonon coupling. Our results are relevant for but not limited only to electron-phonon scattering processes. Other examples could be electron-plasmon, electron-electron, or electron-spin excitation scattering events. In the following, we first briefly recall the relationship between linear conductance and charge susceptibility at equilibrium. Then, we derive the corresponding relationship in the NE conditions and present numerical calculations.

Equilibrium response functions.—Within the linear-response theory of a system at equilibrium [12–14], the current I is related to a frequency-dependent applied bias V via the linear conductance g as $I(\omega) = g(\omega)V(\omega)$. The linear conductance is a response function obtained from the current-density correlation function $g(t) = (ie/\hbar) \times \langle[\hat{I}(t), \hat{N}(0)]\rangle\theta(t)$, where $\hat{N}$ is the total occupancy operator and $\hat{I}$ is the current operator $\hat{I} = ed\hat{N}/dt$. The linear conductance g is directly related to the density-density correlation function $\chi_c(t) = -i\langle[\hat{N}(t), \hat{N}(0)]\rangle\theta(t)$ by the relation $g(\omega) = i\omega\frac{e^2}{\hbar}\chi_c(\omega)$. χ_c is also known as the

charge susceptibility and represents the response function of the charge density modifications δn due to variation of the electrostatic potential δv: $\delta n = \int \chi_c \delta v$ [15]. In the dc limit, one gets a finite linear conductance $g(\omega \to 0)$ when the charge susceptibility goes as $\chi_c(\omega) = f(\omega)/\omega$ with $f(0) \neq 0$. At equilibrium, there is a clear and well-defined relationship between the charge susceptibility and the linear conductance. However, there is no *a priori* reason why such a relation should still hold at nonequilibrium when an applied bias drives the system in a nonlinear regime.

Nonequilibrium charge susceptibility and transport.—We consider a generic system consisting of a interacting central region C, the scatterer of interest (e.g., a molecule or a quantum dot), connected to two electrodes acting as source and drain. The electrodes are noninteracting Fermi seas at their own equilibrium, and there is no direct contact between them. We use nonequilibrium Green's functions to calculate the electric current and charge of the system in NE conditions [16]. The system is under a finite, but not small, applied bias and is assumed to have reached a nonequilibrium steady state which can be described by an effective (pseudo-)equilibrium steady-state density matrix [8,17].

We define the nonequilibrium charge susceptibility χ_c^{NE} in the steady state as the response (not necessarily linear) for the modifications of the total electronic occupancy of the central region $\delta\langle n_C\rangle$ due to the changes in the applied bias δV, i.e., changes in the cause that drives the system out of equilibrium [18]:

$$\chi_c^{\mathrm{NE}}(V) = \frac{\partial\langle n_C^{\mathrm{NE}}\rangle}{\partial V}. \tag{1}$$

The total occupancy $\langle n_C^{\mathrm{NE}}\rangle$ of the central region C is given by the nonequilibrium lesser Green's function as $\langle n_C^{\mathrm{NE}}\rangle = -i\int d\omega \mathrm{Tr}[G^<(\omega)]/2\pi$, where the trace runs over the electronic states in the region C.

We now examine in detail how $\chi_c^{\mathrm{NE}}(V)$ is related to the dynamical conductance $G(V) = dI/dV$. The current at the left L interface between the central region C and the L lead is given by the Meir-Wingreen expression [16]

$$I_L = \frac{ie}{\hbar}\int \frac{d\omega}{2\pi}\,\mathrm{Tr}\{f_L(\omega)[G_C^r(\omega) - G_C^a(\omega)]\Gamma_L(\omega) + G_C^<(\omega)\Gamma_L(\omega)\}, \tag{2}$$

with $\Gamma_L(\omega)/2$ being the imaginary part of the L lead self-energy and $G_C^{r,a,<}$ being the retarded, advanced, and lesser Green's functions of the central region, respectively, and the trace is taken over the electron states of the central region.

By using the properties of a NE steady state, one introduces a nonequilibrium distribution functional f_C^{NE} for the central region as $G_C^<(\omega) = -f_C^{\mathrm{NE}}(\omega)(G_C^r - G_C^a)(\omega)$ [19]. At equilibrium, f_C^{NE} is simply given by the conventional Fermi distribution function. The dynamical conductance $G(V)$ can be written as

$$G(V) = \frac{ie}{\hbar}\int \frac{d\omega}{2\pi}\,\mathrm{Tr}\{[1 - f_L(f_C^{\mathrm{NE}})^{-1}]\partial_V G_C^<\Gamma_L - \partial_V[f_L(f_C^{\mathrm{NE}})^{-1}]G_C^<\Gamma_L\}, \tag{3}$$

which shows a relation between the dynamical conductance and the derivative of the lesser Green's function versus the applied bias $\partial_V G^<$. To show more clearly how $G(V)$ and $\chi_c^{\mathrm{NE}}(V)$ are related to each other, we consider the following simpler system.

A model system.—The model consists of a single electron level in the region C, in the presence of some arbitrary kind of interaction. For the moment, we consider the wideband limit where $\Gamma_L(\omega) = \Gamma$ and that all the potential drop occurs at the left contact. Only the Fermi distribution f_L of the left lead depends explicitly on the bias V via the Fermi level μ_L. Within these conditions, we find a relation between the dynamical conductance G and the nonequilibrium charge susceptibility χ_c^{NE}:

$$G(V)\left(\frac{e}{\hbar}\Gamma\right)^{-1} + \chi_c^{\mathrm{NE}}(V) = \int d\omega\,\partial_V[f_L A_C(\omega)], \tag{4}$$

where $A_C(\omega) = [G_C^a(\omega) - G_C^r(\omega)]/2\pi i$.

For noninteracting systems, the spectral function A_C is independent of the bias; then, $\partial_V A_C(\omega) = 0$. By using the definitions of G and χ_c^{NE} for symmetric contacts and the corresponding nonequilibrium distribution function $f_C^{\mathrm{NE}} = (\Gamma_L f_L + \Gamma_R f_R)/(\Gamma_L + \Gamma_R) = (f_L + f_R)/2$ [20], we find a direct proportionality between G and χ_c^{NE}: $G(V) = \frac{e^2}{\hbar}\Gamma\chi_c^{\mathrm{NE}}(V)/e$ [21]. Beyond the wideband approximation (with symmetric contacts), we obtain the relation $G(V) = \frac{e^2}{\hbar}\Gamma(\mu_L)\chi_c^{\mathrm{NE}}(V)/e$. Hence, the compatibility between the equilibrium and NE approaches implies that $\lim_{\omega\to 0} i\omega\chi_c(\omega) \equiv \Gamma\chi_c^{\mathrm{NE}}(V)$ (within the dc limit of linear response).

For interacting systems, A_C depends on V through the interaction self-energy $\Sigma_{\mathrm{int}}(\omega, V)$. An analytical expression relating G and χ_c^{NE} is more difficult to obtain [22]. However, we show next, from numerical calculations beyond the wideband limit, that there is a clear relationship between $G(V)$ and $\chi_c^{\mathrm{NE}}(V)$ for a model of interaction self-energy.

An application.—For this, we have to make a choice for the interactions in the central region C. The NE charge susceptibility has been briefly studied for a model of electron-electron interaction in the Anderson impurity model at nonequilibrium in [23]. In the following, we consider a model electron-phonon interaction in the central region C [24,25]. Considering such a model permits us to get several different physical effects: the renormalization of the electron level, but also all the phonon replicas (the phonon sideband peaks). So, effectively, we are dealing with a richer model of multielectronic resonances. Such a model includes different inelastic scattering events: those related to charge fluctuations in the electronic resonances (resonant elastic and inelastic transmission) and those

involving off-resonant inelastic scattering by tunneling electrons. However, the relationship derived previously for $G(V)$ and $\chi_c^{\mathrm{NE}}(V)$ is independent of the nature of the interaction (electron-phonon or electron-electron) in the central region C.

In our model, the Hamiltonian for the region C is

$$H_C = \varepsilon_0 d^\dagger d + \omega_0 a^\dagger a + \gamma_0 (a^\dagger + a) d^\dagger d, \qquad (5)$$

where $d^\dagger$ (d) creates (annihilates) an electron in the level ε_0, which is coupled to the vibration mode of energy ω_0 via the coupling constant γ_0. The central region C is connected to two (left and right) one-dimensional tight-binding chains via the hopping integrals t_{0L} and t_{0R}. The corresponding lead $\alpha = L, R$ self-energy is $\Sigma_\alpha^r(\omega) = t_{0\alpha}^2/\beta_\alpha \exp^{ik_\alpha(\omega)}$, with the dispersion relation $\omega = \varepsilon_\alpha + 2\beta_\alpha \cos[k_\alpha(\omega)]$. Here, the imaginary part $\Gamma_\alpha = -2\mathrm{Im}\Sigma_\alpha^r$ is energy-dependent and goes beyond the wideband limit, unless β_α is much larger than any other parameters. At equilibrium, the whole system has a well-defined unique Fermi level μ^{eq}. A finite bias V, applied across the junction, lifts the Fermi levels as $\mu_{L,R} = \mu^{\mathrm{eq}} + \eta_{L,R} eV$. The fraction of potential drop at the left contact is η_L, and $\eta_R = \eta_L - 1$ at the right contact [26], with $\mu_L - \mu_R = eV$ and $\eta_L \in [0, 1]$.

Finally, the electron-phonon interaction is treated at the Hartree-Fock level (first-order diagrams in terms of the interaction) and is incorporated as self-energies $\Sigma_{e\text{-ph}}^{\mathrm{HF},x}(\omega)$ in the nonequilibrium Green's function (x represents the different components: advanced, retarded, greater, and lesser). Self-consistent calculations provide a partial resummation of the diagrams to all orders [24,25].

Within this model, we calculate the dynamical conductance $G(V)$ from Eq. (2) and the NE charge susceptibility $\chi_c^{\mathrm{NE}}(V)$ from Eq. (1) for different sets of parameters. We consider symmetric ($t_{0L} = t_{0R}$) and asymmetric ($t_{0L} \neq t_{0R}$) coupling to the leads, different strengths of coupling to the leads, symmetric and asymmetric potential drops at the contacts, and different transport regimes (off-resonant $\varepsilon_0 \ll \mu^{\mathrm{eq}}$, and resonant $\varepsilon_0 \sim \mu^{\mathrm{eq}}$). We restrict ourselves here to the medium electron-phonon coupling ($0.5 < \gamma_0/\omega_0 < 1$) regime which corresponds to realistic coupling in organic molecules. The strong coupling regime requires higher-order diagrams and more time-consuming calculations [24,25].

Figure 1 shows the NE charge susceptibility $\chi_c^{\mathrm{NE}}(V)$ and the dynamical conductance $G(V)$. We consider a symmetric coupling to the leads ($t_{0L} = t_{0R}$) and an asymmetric potential drop ($\eta_L = 1$). On this scale, both the conductance and the NE charge susceptibility present peaks at an applied bias corresponding to an electronic resonance: a main resonance peak close to full polaron shift-renormalized level $\tilde{\varepsilon}_0 = \varepsilon_0 - \gamma_0^2/\omega_0$ and phonon sideband peaks around $V \sim \tilde{\varepsilon}_0 + n\omega_0$ [27]. In the NE conditions, the charge fluctuates in these electronic resonances whenever the bias window includes $\tilde{\varepsilon}_0 + n\omega_0$. Hence,

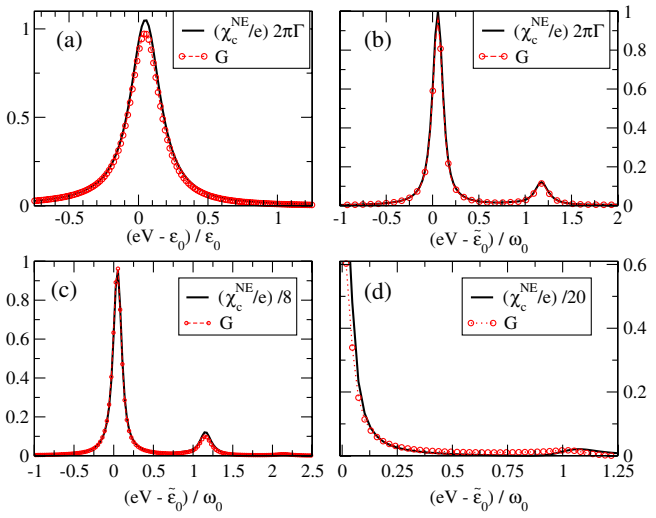

FIG. 1 (color online). Nonequilibrium charge susceptibility χ_c^{NE} (solid lines) and dynamical conductance G (dashed lines) versus applied bias. (a) Noninteraction case, with $\varepsilon_0 = 0.5$ and $\beta_\alpha = 0.7$. (b),(c) With interaction and for different transport regimes ($\beta_\alpha = 2$). From off-resonant to resonant: (b) $\varepsilon_0 = 0.7$, (c) $\varepsilon_0 = 0.5$, and (d) $\varepsilon_0 = 0.15$. χ_c^{NE} is rescaled by $\Gamma = \Gamma_\alpha(\mu^{\mathrm{eq}}) = t_{0\alpha}^2/\beta_\alpha$. On this scale, both χ_c^{NE} and G present the same spectral features: peaks associated with charge fluctuations in the electronic resonances. Calculations are done for symmetric coupling $t_{0\alpha} = 0.15$ and asymmetric potential drops $\mu_L = \mu^{\mathrm{eq}} + eV$ and $\mu_R = \mu^{\mathrm{eq}}$. The other parameters are $\omega_0 = 0.3$, $\gamma_0 = 0.21$, and $\varepsilon_\alpha = 0$. The energy parameters are in eV, and G is in units of $G_0 = e^2/h$.

peaks are obtained in the charge susceptibility $\chi_c^{\mathrm{NE}}(V)$ for these biases. The peaks correspond to elastic ($V \sim \tilde{\varepsilon}_0$) and inelastic ($V \sim \tilde{\varepsilon}_0 + n\omega_0$) resonant scattering processes. For the noninteracting case—see Fig. 1(a)—there is only one resonance at ε_0, and, as demonstrated, χ_c^{NE} and G are related via $\Gamma(\mu_L)$ beyond the wideband limit.

Figure 2 shows that the relationship between χ_c^{NE} and G is robust against our model parameters. It holds for asymmetric coupling to the leads ($t_{0L} \neq t_{0R}$) and different fractions of potential drops at the contacts—see Fig. 2(d). It holds for strong coupling to the leads $t_{0\alpha} \sim \omega_0 > \gamma_0$—see Fig. 2(c)—and beyond the wideband limit—see Fig. 2(b). It also holds when the interaction is modeled only with the Fock diagram—see Fig. 2(a). Therefore, the relationship between χ_c^{NE} and G is not due to the fact that the Hartree self-energy $\Sigma_{e\text{-ph}}^H$ is proportional to $\langle n_C^{\mathrm{NE}} \rangle$ (hence, $\partial_V \Sigma_{e\text{-ph}}^H \propto \chi_c^{\mathrm{NE}}$).

Note that, with potential drops at both contacts, μ_L and μ_R support a fraction of the bias, and the relationship between χ_c^{NE} and G includes terms in $\partial_V f_R$. However, both quantities still present the same features versus applied bias—see Fig. 2(d).

On a smaller energy scale, the conductance also contains physical information for biases around excitation energies which goes beyond resonant transmission. Indeed, the

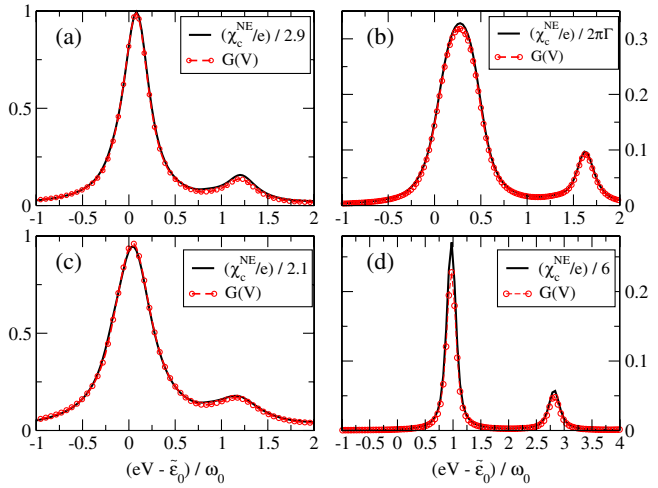

FIG. 2 (color online). Nonequilibrium charge susceptibility $\chi_c^{\rm NE}$ (solid lines) and dynamical conductance G (dashed lines) versus V. Same parameters as in Fig. 1(c), except as otherwise stated. (a) Far beyond the wideband approximation: $\beta_\alpha = 0.7$. (b) Interaction at the Fock level only. (c) Strong coupling to the leads $t_{0\alpha} = 0.30 = \omega_0$. (d) Asymmetric coupling to the leads and potential drop, $t_{0L} = 0.07$, $t_{0R} = 0.15$, $\eta_L = t_{0R}/(t_{0L} + t_{0R}) = 0.681\,82$, and $\varepsilon_0 = 0.70$.

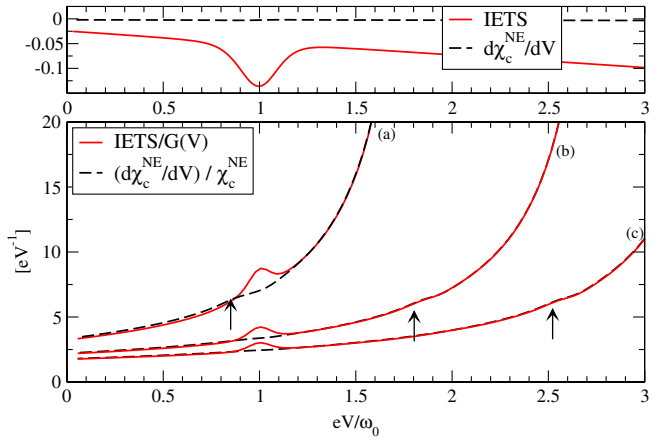

FIG. 3 (color online). Derivative $\partial_V \chi_c^{\rm NE}$ (dashed lines) and IETS signal $\partial_V G$ (solid lines). $\partial_V \chi_c^{\rm NE}$ does not have the peak or dip feature of the IETS at $V \sim \omega_0$. (*Top*) Resonant transport regime. Calculations were done with $t_{0\alpha} = 1.50 \sim \beta_\alpha$, $\varepsilon_0 = 0.0$, and $\gamma_0 = 0.195$. (*Bottom*) Off-resonant regime for different ε_0. $\partial_V \chi_c^{\rm NE}$ and $\partial_V G$ are normalized by $\chi_c^{\rm NE}$ and G, respectively. (a) $\varepsilon_0 = 0.70$, (b) $\varepsilon_0 = 0.99$, and (c) $\varepsilon_0 = 1.20$. The arrows point out the position of the electron resonance at $V = \tilde{\varepsilon}_0 - \omega_0$. Calculations were done with the same parameters as in Fig. 1(b).

conductance also varies at bias thresholds corresponding to other inelastic scattering processes (for example, inelastic electron tunneling). At the bias threshold $V \sim \omega_0$, the conductance increases in the off-resonant transport regime (opening of new conduction channels) or decreases in the resonant transport regime (electron-phonon backscattering). These effects are better seen in the inelastic electron tunneling spectroscopy (IETS) as peaks or dips for the off-resonance or resonant transport regime, respectively [25,28]. The IETS is obtained from the second derivative of the current versus applied bias $d^2I/dV^2 = dG/dV \equiv \partial_V G(V)$. In experiments, the IETS signal is usually given as normalized by the conductance itself or by the current itself. Figure 3 shows the IETS signal, as well as the corresponding variation of the nonequilibrium charge susceptibility versus applied bias $\partial_V \chi_c^{\rm NE}$. One can clearly see a peak feature at $V \sim \omega_0$ in the IETS signal, while $\chi_c^{\rm NE}$ is virtually featureless at the corresponding bias for both the off-resonant and resonant transport regimes. This means that these inelastic tunneling electron-phonon scattering processes (at $V \sim \omega_0$) are not related to charge fluctuations. Instead, the phonon population fluctuates because of phonon emission induced by the tunneling electron. Note that the tiny features pointed out by the arrows in Fig. 3 correspond to tiny peak features in both $\chi_c^{\rm NE}$ and G. They are related to charge fluctuations in the electron resonances at $V = \tilde{\varepsilon}_0 - \omega_0$ (phonon emission by a hole).

Discussion.—We have hence shown that the nonequilibrium charge susceptibility and the dynamical conductance are directly related to each other, although in a different manner than for the equilibrium case. They both present features (peaks) versus the applied bias whenever there are charge fluctuations in the corresponding electronic resonances of the nanojunction.

Therefore, we suggest that measuring both the conductance and the NE charge susceptibility simultaneously in the same experiment is essential in quantum transport. It permits one to identify the nature of the scattering processes involved in the transport, i.e., processes involving charge fluctuation or not. This result is very important for the analysis of the transport properties in complex systems such as large single-molecule junctions and does not involve the presence of a third gate electrode. Although our result is mostly relevant for electron-phonon scattering processes, it is not limited only to these processes. The measurement of the NE charge susceptibility could be performed by measuring the potential drop around a capacitor placed in series with the nanojunctions ($V_{\rm cap} = e\langle n_C\rangle/C_{\rm cap}$). One can then obtain $\chi_c^{\rm NE}(V)$ in a similar way as the dynamical conductance $G(V)$ is obtained from the current by using a lock-in setup.

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