

Generalization and applicability of the Landauer formula for nonequilibrium current in the presence of interactions

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Using nonequilibrium Green's functions (NEGFs), we calculate the current through an interacting region connected to noninteracting leads. The problem is reformulated in such a way that a Landauer-like term appears in the current as well as extra terms corresponding to nonequilibrium many-body effects. The interaction in the central region renormalizes not only the Green's functions but also the coupling at the contacts between the central region and the leads, allowing the total current to be further expressed as a generalized Landauer-like current formula. The general expression for the dynamical functional that renormalizes the contacts is provided. We analyze in detail under what circumstances Landauer-like approaches to the current, i.e., without contact renormalization, are valid for interacting electron-electron and/or electron-phonon systems. Numerical NEGF calculations are then performed for a model electron-phonon coupled system in order to validate our analytical approach. We show that the conductance for the off-resonant transport regime is adequately described by Landauer-like approach in the small-bias limit while for the resonant regime, the Landauer-like approach results depart from the exact results even at small finite bias. The validity of applying a Landauer-like approach to inelastic electron-tunneling spectroscopy is also studied in detail.

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I. INTRODUCTION

Electronic transport through nanoscale systems exhibits many important new features in comparison with conduction through macroscopic systems. In particular, local interactions, such as Coulomb interactions between the electrons and scattering from localized atomic vibrations, become critically important. In crude terms, these effects are more important in nanoscale systems as the electronic probability density is concentrated in a small region of space; normal screening mechanisms are thus ineffective.

It is most useful to have a simple expression for the electronic current or for the conductance of a nanoscale object connected to terminals. This is provided in the form an appealing intuitive physical picture by the Landauer formula,¹ which describes the current in terms of the transmission coefficients of the central scattering region and of distribution functions of the electrons in the terminals. However, in its original form the Landauer formula deals only with noninteracting electrons. This formalism has been used in conjunction with density-functional theory (DFT) calculations for realistic nanoscale systems^{2–12} and has helped tremendously for the qualitative understanding of the transport properties of such realistic systems. The apparent success of such approaches relies on the fact that DFT maps the many-electron interacting system onto an effective noninteracting single-particle Kohn-Sham Hamiltonian suited for the Landauer formalism for transport. However when such a mapping becomes questionable for strongly interacting electron systems, the original Landauer approach has been found to be incomplete and unable to properly take into account the many-body effects.^{13,14}

The Landauer formula has been built upon by Meir and Wingreen¹³ to extend this formalism to a central scattering region containing interacting between particles. It is then ex-

pressed in terms of nonequilibrium Green's functions and self-energies and in the most general cases it does not bear any formal resemblance with the original Landauer formula for the current.^{13,15} Other generalization of Landauer-like (LL) approaches to include interactions and inelastic scattering have been developed, see, for example, Refs. 14 and 16–18.

It is therefore important to know the domain of validity of Landauer-like approaches in comparison to exact current calculations based on nonequilibrium Green's functions for treating electron transport through an interacting region connected to leads at different thermodynamical equilibria. This is the question we address in this paper by following a two-step approach.

First we reformulate Meir and Wingreen's work to once more express the current as the sum of a Landauer-like expression involving a transmission coefficient, plus a non-Landauer-like term arising from the nonequilibrium many-body effects. We further develop our theoretical framework to show that the interaction between particles in the central region not only renormalizes the nonequilibrium Green's functions but also the coupling at the contacts between the central region and the leads. We hence obtain a generalized Landauer-like formula for the current in the same spirit as in Refs. 16, 17, 19, and 20. However, our result for the dynamical functional that renormalizes the coupling at the contacts is more general than the ansatz used in previous studies (Refs. 16, 17, and 19–21). Our result does not imply any constraints on the statistics of the nonequilibrium interacting central region.

Second, we apply our theoretical framework to study a model system in the presence of electron-phonon (electron-vibron) interactions, connected to two noninteracting electron leads at nonequilibrium. We analyze in detail the validity of Landauer-like approaches to describe the conductance

and the inelastic electron-tunneling spectroscopy (IETS) of such a nonequilibrium many-body interacting system.

The paper is organized into two main sections. In Sec. II, we develop our formalism to derive our generalized expression for the current. The implications of this are discussed in Sec. II C. We then apply this formalism to the model system in Sec. III and show the results of numerical calculations.

II. FORMALISM

A. Model system

Following Meir and Wingreen,¹³ we consider a scattering central region (a quantum dot, a molecule, or a nanowire including interaction between particles) which is connected to two (left L and right R) leads. These leads are described by two noninteracting Fermi seas at their own equilibrium, characterized by two Fermi distributions $f_L(\omega)$ and $f_R(\omega)$.

The Hamiltonian of the system is given by

$$\hat{H} = \sum_{\alpha=L,R} \varepsilon_\alpha \hat{c}_\alpha^\dagger \hat{c}_\alpha + \hat{H}_{\text{int}}(\{\hat{d}_n^\dagger\}, \{\hat{d}_n\}; \{\hat{a}_\lambda^\dagger\}, \{\hat{a}_\lambda\}) + \sum_{n,\alpha=L,R} (V_{\alpha,n} \hat{c}_\alpha^\dagger \hat{d}_n + \text{H.c.}), \quad (1)$$

where the summation indices α run over the left and right leads (L, R respectively) and depending on the choice of representation over momentum k or lattice site i index, with $\hat{c}_\alpha^\dagger(\hat{c}_\alpha)$ creating (annihilating) a noninteracting electron, and $\{\hat{d}_n^\dagger\}, \{\hat{d}_n\}$ represent a complete, orthonormal set of states for the interacting electrons in the central region, and $\{\hat{a}_\lambda^\dagger\}, \{\hat{a}_\lambda\}$ represent a set of bosonic degrees of freedom to which the electrons are coupled in the central region. These can be more or less extended phonons in a quantum dot or nanowire, or molecular vibrations (vibrons) in molecules.

There are two main approximations in the Meir and Wingreen approach to transport. The first is to consider that the interactions are localized within the central region. This leads to specific properties for the self-energies used to calculate the nonequilibrium many-body Green's functions within the basis states of the central region only. The self-energies are then obtained as the sum of three contributions: two similar contributions arising from the electronic coupling of the central region to the left and right leads and the third arising from the interaction between particles in the central region.

The second approximation is to consider that the initial correlations die out in the long-time limit, and hence a steady-state regime can be reached. It should be noted that a generalization going beyond the steady-state regime has recently been given in Ref. 22.

B. Nonequilibrium Green's functions and Landauer-like formula for the current

In the steady state, the current I_L flowing at the left contact between the left lead and the central region is expressed in terms of three nonequilibrium Green's functions (the retarded G^r , advanced G^a , and lesser $G^<$ Green's functions) of the dressed interacting central region.¹³

Using the identity $G^>-G^<=G^r-G^a$ and the definition of the leads' self-energies $\Sigma_L^<(\omega)=if_L(\omega)\Gamma_L(\omega)$ and $\Sigma_L^>(\omega)=-i[1-f_L(\omega)]\Gamma_L(\omega)$, for which we recall that $f_L(\omega)$ is the Fermi distribution of the noninteracting left lead and Γ_L is obtained from the imaginary part of the retarded (advanced) self-energy $\Sigma_L^{r(a)}$ arising from the coupling of the central region to the left lead, i.e., $\Gamma_L(\omega)=\mp 2\Im m \Sigma_L^{r/a}(\omega)$, the current I_L is given by²³⁻³⁶

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

where the trace runs over indexes n, m appropriately chosen to represent the electronic states of the central region.

A similar expression can be obtained for the current I_R flowing at the right contact between the right lead and the central region by exchanging the subscript $L \leftrightarrow R$ in Eq. (2). For a current-conserving system, one then has $I_L=-I_R$. The famous result of Meir and Wingreen, [Eq. (6) in Ref. 13], is then obtained by evaluating the symmetrized current, $I=(I_L-I_R)/2$, to give

$$I = \frac{ie}{\hbar} \int d\omega \text{Tr}[[f_L(\omega)\Gamma_L - f_R(\omega)\Gamma_R][G^r(\omega)-G^a(\omega)] + [\Gamma_L(\omega) - \Gamma_R(\omega)]G^<(\omega)]. \quad (3)$$

Now, we can define more explicitly the specific property of the self-energies, namely additivity: $\Sigma^x(\omega)=\Sigma_L^x(\omega)+\Sigma_R^x(\omega)+\Sigma_{\text{int}}^x(\omega)$ where x is any component $x=r, a, >, <$, and the self-energies are defined within the central region by $\Sigma_\alpha^x(\omega)$ for the coupling of the central region to the lead α , and by $\Sigma_{\text{int}}^x(\omega)$ for the interaction between electrons or between electrons and phonons/vibrons. As mentioned in the previous section, the many-body interaction self-energy can be added to the leads' self-energies only because the interactions are localized in the central region. Throughout the paper, we will also use a more compact notation for the leads' self-energy, i.e., $\Sigma_{L+R}^x=\Sigma_L^x+\Sigma_R^x$.

Using the additivity property of the self-energy, and the fact that $G^<(\omega)=G^r(\omega)\Sigma^<(\omega)G^a(\omega)$ in the steady-state regime, the symmetrized current I can be re-expressed as follows:

$$I = \frac{2e}{\hbar} \int \frac{d\omega}{2\pi} \left((f_L-f_R)\text{Tr}[\Gamma_L G^r \Gamma_R G^a] + \text{Tr}\left[(f_L \Gamma_L - f_R \Gamma_R) G^r \frac{i(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<)}{2} G^a\right] + \text{Tr}\left[(\Gamma_L - \Gamma_R) G^r \frac{i \Sigma_{\text{int}}^<}{2} G^a\right] \right). \quad (4)$$

Introducing the nonequilibrium distribution function matrix for the interaction $f_{\text{int}}^{\text{NE}}$ defined from the interaction self-energies as $\Sigma_{\text{int}}^<=-f_{\text{int}}^{\text{NE}}(\Sigma_{\text{int}}^r - \Sigma_{\text{int}}^a)$ (see Appendix A), one can rewrite the symmetrized current I as follows:

$$I = \frac{2e}{\hbar} \int \frac{d\omega}{2\pi} \left((f_L - f_R) \text{Tr}[\Gamma_L G' \Gamma_R G^a] + \text{Tr} \left\{ [(f_L - f_{\text{int}}^{\text{NE}}) \Gamma_L - (f_R - f_{\text{int}}^{\text{NE}}) \Gamma_R] \times G' \frac{i(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<)}{2} G^a \right\} \right). \quad (5)$$

The first term in Eqs. (4) and (5) looks like a LL expression for the current,

$$\begin{aligned} I^{\text{LL}} &= \frac{2e}{\hbar} \int d\epsilon [f_L(\epsilon) - f_R(\epsilon)] T_{\text{eff}}(\epsilon) \\ &= \frac{2e}{\hbar} \int \frac{d\omega}{2\pi} (f_L - f_R) \text{Tr}[\Gamma_L G' \Gamma_R G^a] \end{aligned} \quad (6)$$

with an effective transmission

$$T_{\text{eff}}(\epsilon) = \text{Tr}[\Gamma_L G' \Gamma_R G^a](\epsilon) = \text{Tr}[t^\dagger(\epsilon) t(\epsilon)], \quad (7)$$

which can be interpreted with the intuitive physical picture, as in the original Landauer formulation of electronic transport, in terms of transmission coefficients $t(\epsilon)$ and propagation eigenchannels as defined in Refs. 37 and 38.

The second term in Eq. (5) corresponds to nonequilibrium corrections due to the interaction. It is expressed in terms of $\Sigma_{\text{int}}^{<, >}$ and of the different distribution functions. This term, not automatically small, cannot be recast in the form of extra transmission coefficients as in Refs. 37 and 38, and already indicates in a way the breakdown of the original Landauer formula for the current in the presence of interaction.¹³

One should note also that even if I^{LL} looks like a Landauer formula for the current with an effective transmission $T_{\text{eff}}(\epsilon)$, the interaction between particles is already taken into account in an exact calculation of the Green's functions. In this sense, I^{LL} is not a conventional Landauer current formula for single-particle elastic scattering. The renormalization of the noninteracting reference system is included in the retarded and advanced Green's functions via the corresponding self-energies: $G^{r,a}(\omega) = [g_0^{r,a}(\omega)^{-1} - \Sigma_{L+R}^{r,a}(\omega) - \Sigma_{\text{int}}^{r,a}(\omega)]^{-1}$. Depending on the way the interactions are treated, the renormalization of the Green's functions may even go beyond the quasiparticle description of the interacting system. In any case, the important point is that T_{eff} already contains part of the electron-electron and/or electron-phonon inelastic-scattering processes.

To complete our theoretical framework, we can make a further formal manipulation of the equations for the exact current, as given, for example, by Eq. (5), and end up with a more compact expression for the current which expresses a clear physical result: the interaction renormalizes not only the Green's functions ($G^{r,a}$) but also the coupling at the contacts.

To show this, it is more convenient to consider for the moment the current at only one contact (I_L , for example), though one should not forget that in the steady state the current conservation implies $I_L = -I_R = I$. The compact expression we find for I_L is the following:

$$I_L = \frac{2e}{\hbar} \int \frac{d\omega}{2\pi} [f_L(\omega) - f_R(\omega)] \text{Tr}[\Gamma_L G' Y_R G^a] \quad (8)$$

with the coupling to the right contact Y_R being renormalized as

$$Y_R(\omega) = \Gamma_R(\omega) \Lambda(\omega) \quad (9)$$

and

$$\Lambda(\omega) = 1 + \Gamma_R^{-1} \frac{f_L(\omega) - f_{\text{int}}^{\text{NE}}(\omega)}{f_L(\omega) - f_R(\omega)} i(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<)(\omega), \quad (10)$$

where we recall that $f_{\text{int}}^{\text{NE}}(\omega)$ is the nonequilibrium statistical distribution for the many-body interactions as defined in Appendix A.

Equations (5) and (8)–(10) [see also Eq. (B3) in Appendix B] represent the principal formal results of this paper. They imply that for an interacting central region, one can always express the current in a generalized Landauer-like formula in which not only the retarded and advanced Green's functions are renormalized by the interaction but also the coupling at the contacts, as similarly found in Refs. 16, 17, and 19–21. This generalized formula needs to be contrasted with the more conventional Landauer-like formula (6) in which the contacts of the central region with the leads are not renormalized by the interaction.

Our expressions (8)–(10) are valid for any kind of interaction localized in the central region and generalize the results of the previous studies (Refs. 16, 17, and 19–21) because they do not imply any restrictions to the nonequilibrium statistics of the many-body interacting central region as we explain in detail in Appendix B.

Finally, one recovers the more conventional Landauer-like formula (with no correction factors or equivalently with no renormalization of the contact couplings) when the quantity $[\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<]$ vanishes, as can be clearly seen from Eqs. (5) and (8)–(10). In the next section, we discuss in detail the conditions for which this can happen. Note that the condition $[\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<] = 0$ does not necessarily imply that $\Sigma^{a,r}(\omega) = 0$ as well. Hence, even when the transport is well described by a Landauer-like formula $\{[\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<] = 0\}$, normalization effects still occur and the transport is dominated by single-quasiparticle scattering.

C. Discussion

Clearly whenever $(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<)(\omega) = 0$, there is no renormalization at the contact, and the current is simply given by I^{LL} . This may happen in two cases: either $\Sigma_{\text{int}}^{<, >} = 0$ for all ω or only within finite range(s) of ω . In the latter case, the relevant range of ω for which $(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<) = 0$ should be included within the bias window defined by the two Fermi levels μ_L and μ_R at nonequilibrium.

In order to understand how and why the quantity $\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<$ can vanish for an interacting system, let us first come back to the definition of the lesser and greater self-energies. These are specific components (projections onto the real time axis) of the more general self-energy $\Sigma_{\text{int}}(\tau, \tau')$ with times τ, τ' defined on the Keldysh time-loop contour.^{35,39–41} Within the Keldysh approach, the lesser $<$ (greater $>$) components

of $\Sigma_{\text{int}}(\tau, \tau')$ imply that the times τ/τ' are located on the forward/backward (backward/forward, respectively) time-ordered branch. $\Sigma^{<,>}(\omega)$ is simply the Fourier transform of $\Sigma_{\text{int}}^{<,>}(t, t')$ in the limit of the steady-state regime where any quantity depends only on the time difference $X(t, t') = X(t - t')$.

First let us examine the first case: why would a self-energy have no lesser or greater components? For the so-called irregular self-energies,⁴² we have the condition $\Sigma(\tau, \tau') = \hat{\Sigma}(\tau) \delta(\tau - \tau')$. The self-energies for the interaction are instantaneous (local) in time. Hence they cannot have lesser or greater components since the times have to be on the same time-loop branch. This condition of locality in time corresponds to two classes of physical effects. First when the self-energies describe one-particle potentials due to electron-electron or electron-phonon interaction, in other words it corresponds to the Hartree-Fock approximation for electron-electron interaction and to only the Hartree-type approximation for electron-phonon interaction and second when the self-energies correspond to the so-called initial correlations which contain all contributions singular in time (see, for example, Refs. 43 and 44).

There is also another class of problems for which there are no lesser or greater components of the self-energy. It is when the exchange and correlation effects for interacting electron systems are represented by an effective potential $v_{xc}(r, t) = \delta A_{xc}[n]/\delta n(r, t)$ being obtained from an exchange-correlation action functional $A_{xc}[n]$ of the electron density $n(r, t)$. To this potential will correspond an effective self-energy that is local in both space and, more importantly, in time,⁴⁵ hence with no lesser and greater components for a generalization onto the Keldysh contour.

In effect, any method which maps an interacting electron/phonon system onto an effective one-particle (quasiparticle) scheme as, for example, in density-functional-based technique (DFT, TDDFT) or other mean-field approaches, will end up with no lesser and greater components for the corresponding self-energy describing the interaction. Hence a Landauer-like approach to the transport is entirely appropriate for such methods.^{2,3} However, the mapping onto a one-particle scheme may not describe well strongly correlated electronic systems, $A_{xc}[n]$ being amenable to approximation, or is simply not possible in the general case of electron-phonon interaction.

Now, let us turn to the second case: the interaction spectral density $\Im m \Sigma_{\text{int}}^r = i(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<)/2$ vanishes for one or more (connected) ranges of ω values. If this gap in $\Im m \Sigma_{\text{int}}^r$ is enclosed within the bias window then again, the current I will be determined only by the Landauer-like term I^{LL} .

As will be shown in detail below from numerical calculations for a model system, such a gap in $\Im m \Sigma_{\text{int}}^r$ may exist in special cases of electron-phonon interaction. The gap in $\Im m \Sigma_{\text{int}}^r$ is then usually located around the Fermi level at equilibrium, and around the Fermi levels for the nonequilibrium cases at low applied bias only. These cases correspond to the regime studied by Imry *et al.*¹⁸ who derived a Landauer-like inelastic transmission for interacting electron-phonon systems and argued that the Landauer picture is still valid in the presence of interaction as long as multiparticle processes can be neglected.

For electron-electron interactions, the situation is somewhat different. Even if collective excitations such as plasmons present some qualitative bosonic analogy to phonons (there is a peak in the self-energy around the plasmon energy—as for the e-ph self-energy—and no much interaction spectral density elsewhere), there is always however a nonzero contribution to the self-energy coming from the continuum of electron-hole excitations.

The main difference from the electron-phonon interaction is that the phonon frequency ω_0 imposes a restricted energy scale on the interaction while for electron-electron interaction all energy scales are available, making the corresponding interaction self-energy not vanishing, except in an infinitely small energy window around the Fermi energy at equilibrium. So, in principle, Landauer-like approaches for interacting electron systems are not valid for interactions treated beyond the mean-field/density-functional-based approximations.¹⁴

Finally, one should note that the second term in Eq. (5) for the current or the second term in the renormalization function $\Lambda(\omega)$ in Eq. (10) involves the quantity $(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<)$ which is, in a series expansion of the interaction, proportional to the powers of the coupling constant(s) characterizing the interaction. In the limit of weak interactions, these terms represent small corrections to the Landauer-like current expression, and also give small contributions in the renormalization of both the Green's functions and the coupling at the contacts. Hence in the limit of weak interactions, conventional Landauer-like approaches could be confidently used and corrected by using perturbation theory for the interaction.^{46–48}

Now we turn to present numerical calculations for a model system including electron-phonon interactions to illustrate our previous analysis. We compare results obtained from the exact current expression (5) and (8) with the current derived from the Landauer-like formula (6), and by using different levels of approximations for the Green's functions.

III. APPLICATION FOR INTERACTING ELECTRON-PHONON MODEL SYSTEMS

In this section, we study in detail the validity of Landauer-like approaches for an interacting model system connected to two noninteracting electron reservoirs at nonequilibrium. We concentrate on a model of electron-phonon interaction with the simplest version of the Hamiltonian [Eq. (1)] for the central part: a single electron level coupled to a single vibration mode—the single-site single-mode (SSSM) model,⁴⁹ which has also been considered in previous studies.^{23–34,36} We also briefly describe below how to calculate the NEGF from this model Hamiltonian; the full theoretical details can be found elsewhere.⁴⁹

We then apply our NEGF technique to the calculations of the transport properties of the junction around equilibrium and out of equilibrium, and thus for different transport regimes. We analyze in detail if and when Landauer-like approaches can provide a good description of the transport properties in comparison to an exact calculation.

A. Model Hamiltonian for electron-phonon coupling

The Hamiltonian for the central region for the SSSM model is then given by

$$H_C = \varepsilon_0 d^\dagger d + \hbar \omega_0 a^\dagger a + \gamma_0 (a^\dagger + a) d^\dagger d, \quad (11)$$

where one electronic level ε_0 and one vibration mode of energy ω_0 are coupled together via the coupling constant γ_0 .

Furthermore we choose a simple model for the structure of the leads, which provides analytical results for the corresponding surface Green's functions but, in principle, there is no particular restriction to be applied to the model or dimensionality of the leads. So in the following, the left L and right R leads are described by two noninteracting one-dimensional semi-infinite tight-binding chains,

$$\begin{aligned} H_L &= \sum_{i=-1}^{-\infty} \varepsilon_L c_i^\dagger c_i + \beta_L (c_i^\dagger c_{i-1} + \text{c.c.}), \\ H_R &= \sum_{i=+1}^{+\infty} \varepsilon_R c_i^\dagger c_i + \beta_R (c_i^\dagger c_{i-1} + \text{c.c.}). \end{aligned} \quad (12)$$

This model provides us with analytical expressions for the matrix elements of the leads' Green's functions at the terminal sites,⁵⁰

$$g_{0\alpha}^r(\omega) = e^{ik_\alpha(\omega)} / \beta_\alpha \quad (13)$$

with $\omega = \varepsilon_\alpha + 2\beta_\alpha \cos k_\alpha(\omega)$, giving rise to semielliptic density of states of the terminal lead sites connected to the central region.

The expression for the coupling of the central region to the L and R leads is then given by

$$V_{LC} + V_{CR} = \sum_{\alpha=L,R} t_{0\alpha} (c_\alpha^\dagger d + d^\dagger c_\alpha) \quad (14)$$

with hopping integrals $t_{0\alpha}$ and $c_{\alpha=L} = c_{i=-1}$, $c_{\alpha=R} = c_{i=+1}$.

B. Nonequilibrium Green's functions for electron-phonon coupled system

We use a NEGF technique to calculate the properties of the system in a similar manner to previous studies.^{23–34,36} The details of our NEGF calculations are described in detail in Ref. 49 but we briefly summarize our application of them here.

The Green's functions are calculated via Dyson-type equations for the retarded and advanced Green's functions $G^{r,a}(\omega)$,

$$G^{r,a}(\omega) = g_C^{r,a}(\omega) + g_C^{r,a}(\omega) \Sigma^{r,a}(\omega) G^{r,a}(\omega), \quad (15)$$

where $g_C^{r,a}$ is the noninteracting Green's function for the isolated central region.

For the greater $G^>(\omega)$ and lesser $G^<(\omega)$ Green's functions, we use a quantum kinetic equation of the form

$$G^{>,<} = (1 + G' \Sigma^r) g_C^{>,<} (1 + \Sigma^a G^a) + G' \Sigma^{>,<} G^a. \quad (16)$$

Here $\Sigma^x(\omega)$, ($x=r, a, >, <$) is a total self-energy consisting of a sum of the self-energies from the constituent parts of the system,

$$\Sigma^x(\omega) = \Sigma_L^x(\omega) + \Sigma_R^x(\omega) + \Sigma_{\text{int}}^x(\omega). \quad (17)$$

The leads' self-energies $\Sigma_{L+R}^x(\omega) = \Sigma_L^x(\omega) + \Sigma_R^x(\omega)$ arising from the noninteracting leads $\alpha=L, R$ are given by

$$\Sigma_\alpha^r(\omega) = t_{0\alpha}^2 g_{0\alpha}^r(\omega) = [\Sigma_\alpha^a(\omega)]^*,$$

$$\Sigma_\alpha^<(\omega) = -2i\Im m[\Sigma_\alpha^r(\omega)] f_\alpha(\omega),$$

$$\Sigma_\alpha^>(\omega) = -2i\Im m[\Sigma_\alpha^r(\omega)] [f_\alpha(\omega) - 1], \quad (18)$$

where $g_{0\alpha}^r$ is given by Eq. (13) and f_α is the Fermi-Dirac distribution for lead α , with Fermi level $\mu_\alpha = \mu^{\text{eq}} + \eta_\alpha eV$ and temperature T_α . At equilibrium, the whole system has a single and well-defined Fermi level μ^{eq} . Out of equilibrium, a finite bias is applied throughout the junctions. Within our model Hamiltonian, the fraction of electrostatic potential drop at the left contact is $\eta_L = \pm \eta_V$ and $\eta_R = \mp (1 - \eta_V)$ at the right contact,⁵¹ hence $\mu_L - \mu_R = eV$ is indeed the applied bias, and $\eta_V \in [0, 1]$.

The self-energy for the interaction in the central region, $\Sigma_{\text{int}}(\omega)$, is obtained from a nonequilibrium many-body perturbation expansion⁴⁹ of the electron-phonon coupling term in the Hamiltonian, Eq. (11). As for a conventional many-body perturbation expansion, the self-energy is associated with a series of Feynman diagrams for the interaction. In the current work, we consider only the lowest-order diagrams, i.e., the Born approximation (BA) or equivalently the Hartree-Fock approximation.^{23,24,26,28–34,36,52,53} The exact expressions for Σ_{int}^x , ($x=r, a, >, <$) at the Hartree-Fock level and beyond are given in Ref. 49 and we do not reproduce them here.

C. Numerical results

We divide the calculations into two different types of transport regimes. The first of these is when either $\varepsilon_0 \ll \mu^{\text{eq}}$ or $\varepsilon_0 \gg \mu^{\text{eq}}$, known as the off-resonant regime. It corresponds to a poorly conducting junction (i.e., semiconductor-like or insulator-like behavior) dominated by strong tunneling at low bias. The second transport regime is when $\varepsilon_0 \sim \mu^{\text{eq}}$ and known as the resonant transport regime. This regime corresponds to a good, metallic-like, conducting junction.

We will see below that depending on the nature of the transport regime, the Landauer-like approaches may be sufficient, under certain conditions, to describe the conductance $G(V) = dI/dV$ or the IETS properties of the junctions.

1. Self-energy quantity ($\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<$)

As discussed in Sec. II C, the quantity $(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<)$ plays the key role in determining whether or not the Landauer-like approaches are valid. This quantity is plotted in Fig. 1 for the off-resonant transport regime and for intermediate electron-phonon coupling strength. Calculations were performed self-consistently using the lowest-order electron-phonon diagrams, i.e., within the conventional self-consistent BA (SCBA).⁴⁹ By definition $\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^< = \Sigma_{\text{int}}^r - \Sigma_{\text{int}}^a$ is a purely imaginary function for conserving approximations.^{54–57} It presents features (peaks) corresponding to the excitations of the system. The features obtained for nonequilibrium conditions, especially when real excitations can be created in the system (applied bias $V \geq \omega_0$), are strongly different than the features obtained for equilibrium (no applied bias $V=0$).

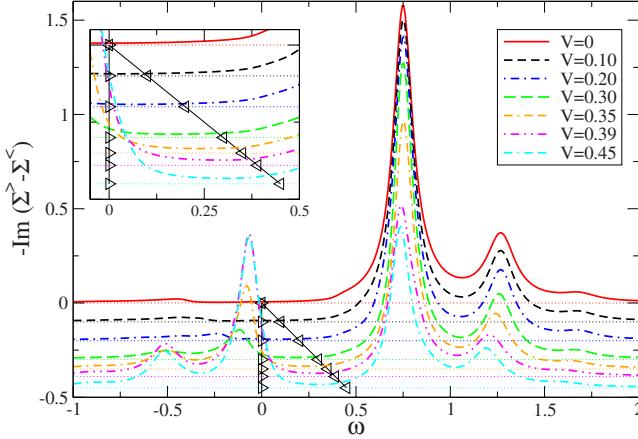


FIG. 1. (Color online) The $(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<)(\omega)$ quantity for the SSSM model within NEGF-SCBA for off-resonant transport regime and intermediate electron-phonon coupling strength $\gamma_0/\omega_0=0.65$. The curves are obtained for different applied bias $eV=\mu_L-\mu_R$, and are offset vertically for clarity. The applied bias is also shown and given by the chemical potentials of the left μ_L (left-pointing arrows) and right μ_R (right-pointing arrows) leads, respectively. The other parameters are $\varepsilon_0=+0.5$, $\omega_0=0.4$, $\gamma_0=0.26$, $t_{0L,R}=0.2$, $T_{L,R}=0.011$, $\eta=0.025$, $\eta_V=1$. At zero and low bias (here $V \leq 0.25$), the interaction self-energies difference $(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<)$ is zero within the bias window, and hence the current is given by a Landauer-like formula.

At zero and low bias ($V \leq 0.6\omega_0$ for the set of parameters used in Fig. 1), the difference between the interaction self-energies $(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<)$ is virtually zero within the bias window $[\mu_R, \mu_L]$. This means that the current in Eq. (5) is effectively given only by the first term I^{LL} , and hence Landauer-like approaches are sufficient to describe the transport properties of the system for such biases.

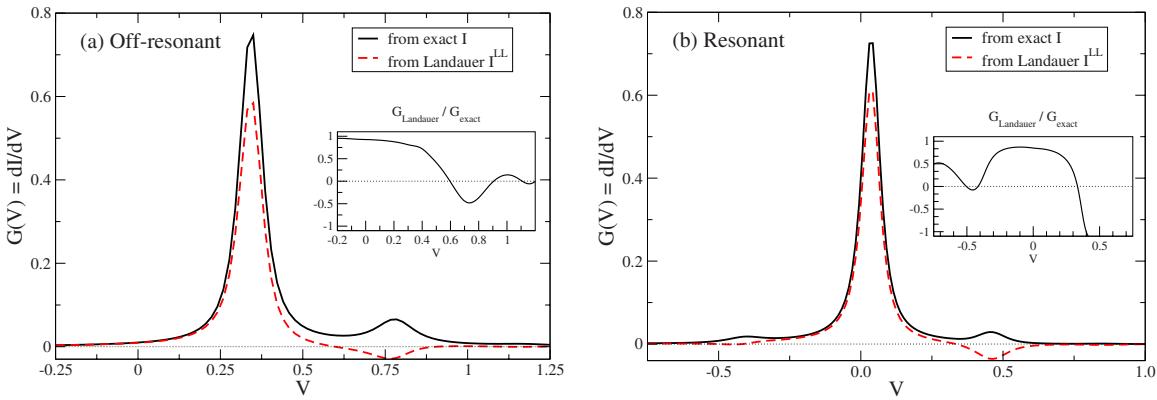


FIG. 2. (Color online) Dynamical conductance dI/dV from the exact expression for the current $I(V)$ and the corresponding Landauer-like current $I^{\text{LL}}(V)$. Green's functions calculations are performed self-consistently for the electron-vibron coupling within the Born approximation and for intermediate coupling $\gamma_0/\omega_0=0.65$. In the off-resonant case (left panel, $\varepsilon_0=+0.5$), there is a good agreement between the two conductances for the linear regime and for low biases, i.e., when $(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<)(\omega) \sim 0$ as shown on Fig. 1. Hence Landauer-like approaches are valid in the low-bias regime for off-resonant transport. For larger biases ($V > 0.5$), dI^{LL}/dV gives unphysical negative conductance values. In the quasiresonant case (right panel, $\varepsilon_0=+0.2$), only the linear conductance is well reproduced by I^{LL} . Large deviations of dI^{LL}/dV from the exact result occur at small applied bias. Hence the Landauer-like formula for nonequilibrium current is not valid for the quasiresonant case. The other parameters for the calculations are the same as used in Fig. 1: $\omega_0=0.4$, $\gamma_0=0.26$, $t_{0L,R}=0.2$, $\eta_V=1$. The insets show the ratio of the conductances calculated with the exact and the corresponding Landauer-like current.

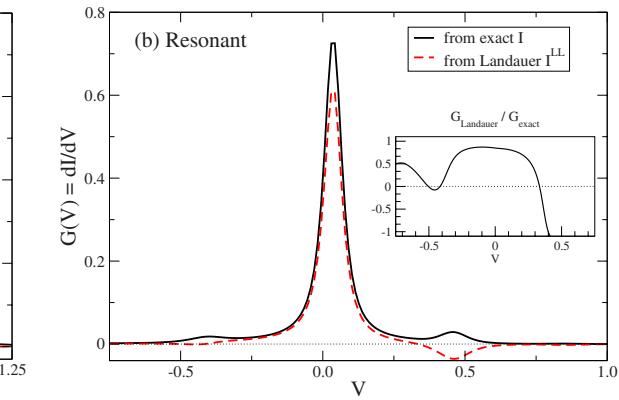
2. Conductance and inelastic electron-tunneling spectroscopy

In order to analyze in detail the different contributions to the conductance and the conditions for which the Landauer-like approaches can be valid, we have performed calculations for the current within different levels of approximation. In the following, we consider four different kinds of approximation: first, in the absence of interaction, the current is obtained from the noninteracting Green's functions $G_0^{r,a}(\omega)=[g_0^{r,a}(\omega)^{-1}-\Sigma_{L+R}^{r,a}(\omega)]^{-1}$ and corresponds to the original Landauer formulation $I^{\text{LL}}[G_0]$ calculated with Eq. (6).

Second, the Landauer current $I^{\text{LL}}[G_0]$ can be corrected to include the interaction effects at the lowest order of the coupling parameters, as in perturbation theory extended to nonequilibrium conditions. This is done by calculating the current in Eq. (4) using only the noninteracting Green's functions $G_0^{r,a}$ in the first term of Eq. (4) and in the evaluation of the interaction self-energies $\Sigma_{\text{int}}^{<,>}[G_0]$; we denote this current by $I_{\text{perturb}}=I^{\text{LL}}[G_0]+\Delta I[\Sigma_{\text{int}}(G_0)]$.

Then the last two kinds of approximations include full renormalization effects in the Green's functions. The first of these corresponds to a nonself-consistent BA calculation using the self-energies $\Sigma_{\text{int}}[G_0]$ to renormalize the Green's functions as follows: $G_{\text{BA}}^{r,a}(\omega)=\{G_0^{r,a}(\omega)^{-1}-\Sigma_{\text{int}}^{r,a}[G_0(\omega)]\}^{-1}$. These Green's functions are then used to calculate the current $I[G_{\text{BA}}]=I^{\text{LL}}[G_{\text{BA}}]+\Delta I[\Sigma_{\text{int}}(G_0)]$. Finally, the last approximation, correspond to a fully self-consistent renormalization SCBA calculation performed as described in Ref. 49, from which we obtain the exact current $I[G_{\text{SCBA}}]=I^{\text{LL}}[G_{\text{SCBA}}]+\Delta I[\Sigma_{\text{int}}(G_{\text{SCBA}})]$.

The dynamical conductance $G(V)$ is obtained as usual from the first derivative of the current versus the applied bias $G(V)=dI/dV$. Typical examples for both the off-resonant and resonant transport regimes are shown in Fig. 2 where we compare the conductance obtained from the exact current with the Landauer-like current $I^{\text{LL}}(V)$ using full SCBA calculations.



For the off-resonant transport regime [Fig. 2(a)], which is dominated by strong tunneling at low biases, there is a good agreement between the exact conductance and the Landauer-like conductance for both the linear regime and the nonlinear regime at low biases, i.e., when $(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<)(\omega) \sim 0$ (as shown on Fig. 1). Hence in this case, Landauer-like approaches are valid to describe the tunneling transport properties in the low-bias regime for off-resonant transport. However, strong discrepancies between the two conductances occur for biases around the first renormalized electronic resonance $V \sim 0.3$, even before real excitations of the phonon mode are available. For even larger biases, ($V \geq 0.5$), dI^{LL}/dV gives unphysical negative conductance values.

For the quasiresonant transport regime [Fig. 2(b)], where the renormalized electronic resonance $\epsilon_0 - \gamma_0^2/\omega_0 \sim 0$ is close to the Fermi level at equilibrium, only the linear conductance is well reproduced by I^{LL} . This is essentially due to the fact that within conservation approximations for the self-energies, the linear conductance is not renormalized by the interaction,^{58,59} as we have also shown in detail in the appendix of Ref. 49. However large deviations of the Landauer conductance from the exact result occur quickly at small applied bias. Hence Landauer-like approaches for nonequilibrium current are not valid for the quasiresonant case, and probably not as well for the resonant case.

We now turn to the IETS which gives information about the selective excitation of the system. The IETS is usually obtained from the second derivative of the current with respect to the applied bias d^2I/dV^2 . The IETS curves present features, peaks or dips,²⁵ at biases corresponding to the energy of a specific excitation, in our case to the energy of one or several excitations of the vibration mode $n\omega$. Being the derivative of the conductance, the IETS curves also present features at biases corresponding to peaks in the conductance (see, for example, Fig. 2).

We have found that in order to get a better aspect ratio for the IETS features corresponding to phonon excitations, it is more convenient to normalize the IETS curves by the dynamical conductance, i.e., $[d^2I/dV^2]/[dI/dV] = d/dV \ln G(V)$. Typical examples for both the off-resonant and resonant transport regimes are shown in Figs. 3 and 4, respectively.

We compare the IETS signals obtained from the four approximations used to calculate the current: I_{Landauer} for the Landauer current of the noninteracting system, I_{perturb} for the Landauer current corrected by first-order perturbation theory for the electron-phonon coupling, and full renormalization within a nonself-consistent and self-consistent scheme $I[G_{\text{BA}}]$ and $I[G_{\text{SCBA}}]$ from which the corresponding Landauer-like contribution can be extracted.

We first comment on the results obtained for the off-resonant case, Fig. 3. As expected, the original Landauer approach does not provide any feature at the bias $V = \omega_0$ since there is no interaction. The IETS signal obtained from the fully self-consistent calculations shows however two features (peaks in the case of strong tunneling regime at low bias^{25,52}) at biases $V = \omega_0$ and $V = 2\omega_0$. They correspond to inelastic processes involving the excitation of one and two vibration modes, respectively.

The rising background of the curves for bias $V \geq 1.2$ corresponds to the feature associated with the main resonance in

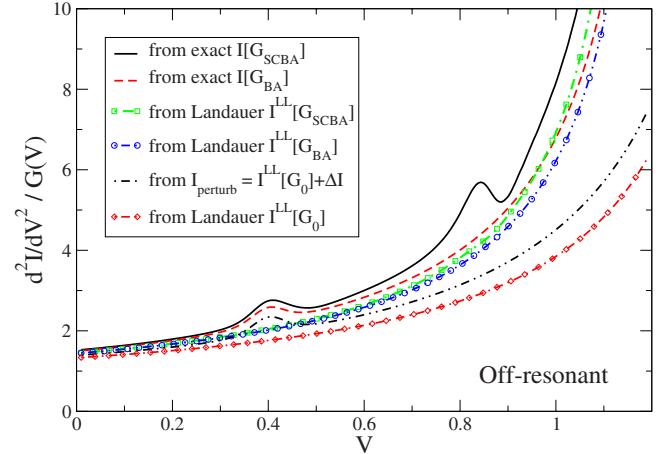


FIG. 3. (Color online) IETS signal d^2I/dV^2 , normalized by dynamical conductance $G(V) = dI/dV$, obtained from the exact expression for the current $I(V)$ and the corresponding Landauer-like term $I^{\text{LL}}(V)$. Calculations are performed for the off-resonant transport regime and the strong-coupling limit $\gamma_0/\omega_0 = 0.875$ using different level of approximations for the Green's functions, as explained in the legend (see main text for detail). The inelastic vibron excitations are present in the IETS derived from the exact $I(V)$ and are located at integer multiples of the vibron energy ω_0 . They give a positive contribution to the baseline, as expected for opaque junctions (insulator/semiconductor-like behavior). However, this signal is absent in the IETS derived from the Landauer-like term $I^{\text{LL}}(V)$. This implies that even with normalized Green's functions the Landauer approaches are not able to correctly reproduce the inelastic features in IETS for the off-resonant regime. The parameters for the calculations are $\epsilon_0 = +1.5$, $\omega_0 = 0.4$, $\gamma_0 = 0.35$, $t_{0L,R} = 0.11$, $T_{L,R} = 0.011$, $\eta = 0.02$, $\eta_V = 1$.

the conductance (see the corresponding main central peak in the conductance curves in Fig. 2). The IETS curve rises at lower bias for the exact calculation compared to the calculations for the noninteracting system, simply because the exact calculations include a full renormalization of the electronic level ϵ_0 . Such a renormalized level is then shifted toward lower energy by the full dynamical polaron shift.^{49,60}

It is interesting to note that the first-order perturbation correction to the Landauer current, given by the second term in Eq. (5) when evaluated from G_0 only, provides not only a qualitatively good feature in the IETS signal at $V = \omega_0$ but also a partial renormalization of the electron resonance. That is, the background of the curve rises faster than for the noninteracting case, and this corresponds to a shift of the electron resonance toward lower energy by a partial polaron shift.

Calculations performed nonself-consistently provide a partial renormalization of the electron resonance, however this is closer to the exact result than that obtained from perturbation theory. Additionally, the corresponding IETS signal also shows only a feature at $V = \omega_0$ as for perturbation theory but its general aspect is again closer to the exact result.

Another important result of our calculations is that the IETS signal calculated from only the Landauer-like term in the current $I[G_{\text{BA}}]$ and $I[G_{\text{SCBA}}]$ does not contain any features at $V = \omega_0$ or $V = 2\omega_0$, as shown in Fig. 3. Although the IETS has the correct rising background and shows the cor-

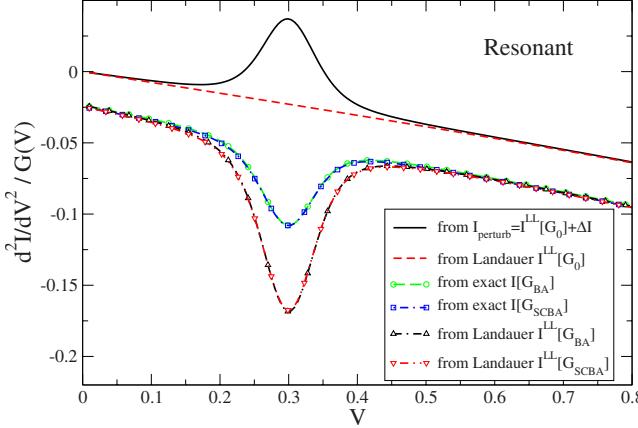


FIG. 4. (Color online) IETS signal d^2I/dV^2 , normalized by $G(V)$, obtained from the exact current $I(V)$ and from the corresponding Landauer-like $I^{LL}(V)$ term. Calculations are for the resonant transport regime, intermediate electron-vibron coupling $\gamma_0/\omega_0=0.65$ and strong coupling to the leads. The different approximations used to calculate the Green's functions are shown in the legend (see main text for detail). The inelastic vibron excitation is present in the IETS signal derived from the exact $I(V)$ and is located around the vibron energy ω_0 . It corresponds to a negative contribution to the baseline, as expected for mostly transparent junctions (metallic-like behavior). Interestingly, this feature is also present in the IETS derived from $I_{LL}(V)$ in contrast to what is obtained for the off-resonant case. Hence, for resonant transport, it seems that Landauer-like approach can reproduce the inelastic IETS features at $V=\omega_0$. The parameters for the calculations are $\varepsilon_0=0$, $\omega_0=0.3$, $\gamma_0=0.195$, $t_{0L,R}=1.50$, $T_{L,R}=0.011$, $\eta=0.025$, $\eta_V=1$.

rect corresponding renormalization of the electronic level, it fails to correctly reproduce the features associated with inelastic processes. Hence in the off-resonant transport regime the renormalization (self-consistent or not) of the Green's functions $G^{r,a}$, from which the Landauer-like current is obtained, does not contain the appropriate physical information to correctly describe the corresponding IETS signal. However, as we have seen above, it is good enough to describe the overall behavior of the conductance at low biases.

Now we turn to the resonant transport regime, and check if the trends obtained for the off-resonant regime hold here as well. It should be noted that in the following calculations, we have considered the resonant transport regime in the case of strong coupling of the central region to the leads, $t_{0L,R} \sim \beta_{L,R}$. We are then dealing with an almost homogeneous one-dimensional system with metallic-like behavior at equilibrium in which the propagating electrons are coupled locally to a single localized vibration mode. We have chosen this somewhat peculiar regime so that the features in the IETS signal associated with the inelastic processes are not “distorted” by the features associated with resonantlike transport. In other words, the background of the IETS signal around the excitation energies is fairly flat.

The corresponding IETS curves are shown in Fig. 4 for the different kinds of approximation used to calculate the current. As expected, the IETS signal calculated for the noninteracting system does not show any feature at $V=\omega_0$

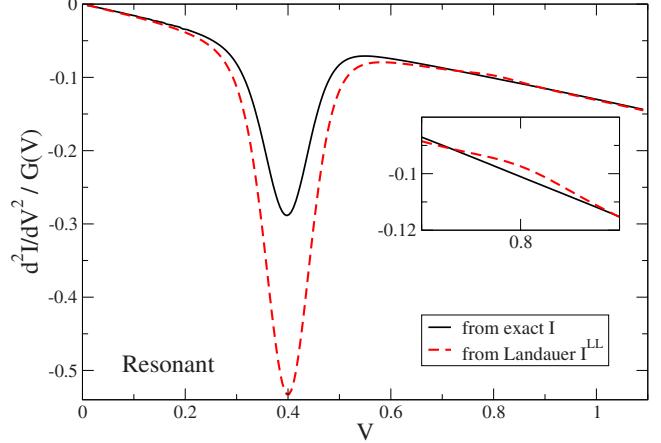


FIG. 5. (Color online) IETS signal d^2I/dV^2 , normalized by $G(V)$, obtained from the exact current expression $I(V)$ and from the corresponding Landauer-like $I^{LL}(V)$ term. The Green's function calculations are performed self-consistently for the resonant case and stronger coupling ($\gamma_0/\omega_0=0.875$) than in Fig. 4. Unexpectedly, even for such a strong coupling, there is no feature in the IETS signal at $V=2\omega_0$ obtained from the exact current. The Landauer-like current provides a small feature at $V=2\omega_0$ with however a spurious positive contribution to the baseline. The other parameters are $\varepsilon_0=0$, $\omega_0=0.4$, $\gamma_0=0.35$, $t_{0L,R}=1.40$, $T_{L,R}=0.015$, $\eta=0.017$, $\eta_V=1$.

while the IETS signal obtained from fully self-consistent calculations show a dip at $V=\omega_0$. Such a negative contribution to the baseline is to be expected in the case of good conductors^{24,25} for which electron-phonon coupling is associated with electron backscattering. The results obtained from nonself-consistent renormalization are very similar to the exact results. Interestingly, the result obtained from perturbation theory gives a feature in the IETS signal at the right bias but however with the wrong sign.

It appears that in all the cases we have studied, first-order perturbation theory always gives a positive contribution (i.e., a peak) to the IETS which is generally incorrect. Indeed it has been shown that the inelastic features of the IETS can be both peaks or dips^{24,25,36,61} depending on the nature of the conductor and essentially on all the parameters describing the system.³⁶

The most interesting result shown in Fig. 4 is that the IETS signal obtained from only the Landauer-like term in the current $I[G_{BA}]$ and $I[G_{SCBA}]$ shows the appropriate feature at $V=\omega_0$. Hence for the resonant transport regime with strong coupling to the leads, the renormalization of the Green's functions from which the Landauer-like current is derived is good enough to describe the IETS signature of the inelastic process at the lowest excitation energy.

Finally, we want to comment on the features in the IETS observed at $V=2\omega_0$, which should correspond to the excitation of two vibron modes by the injected nonequilibrium charge carriers. Such a feature is observed in the rising background of the IETS in the strong tunneling regime (off-resonant transport regime) (Fig. 3); however this feature is clearly absent in the resonant transport case. Even in the case of strong electron-vibron coupling, shown in Fig. 5, there is basically no feature at $V=2\omega_0$ in the IETS signal derived

from the exact expression of the current, although there is a small feature in the IETS signal derived from the Landauer-like current expression. The absence of such a feature at $V=2\omega_0$ for resonant transport is in agreement with previous studies (Refs. 24 and 62). However at the moment there is no satisfactory physical explanation for the absence of such a feature at $V=2\omega_0$. We postulate that such an absence may be due to the partial resummation of the electron-phonon diagrams in the many-body Green's functions. As we have already shown in Ref. 49, higher-order diagrams for the interaction (beyond Hartree-Fock/Born approximation) play a very important role in correctly describing the properties of the electron-phonon coupled system, even at intermediate electron-phonon couplings.

IV. CONCLUSIONS

In this paper, we have readdressed the problem of the breakdown of Landauer-like approaches for electronic transport in the presence of many-body interactions. Starting from the original work of Meir and Wingreen,¹³ we have once more expressed the current as the sum of a Landauer-like expression (I^{LL}) involving the concept of single-particle transmission probabilities plus a non-Landauer-like term arising from the nonequilibrium many-body effects. We have further developed our theoretical framework to show that the interaction in the central scattering region renormalizes not only the nonequilibrium Green's functions but also the coupling at the contacts between the central region and the leads. We have hence obtained a new form of generalized Landauer-like formula for the current, Eqs. (8)–(10), in a similar way to Refs. 16, 17, 19, and 20. However our result for the dynamical functional that renormalizes the coupling at the contacts is more general than the Ng ansatz²¹ used in Refs. 16, 17, 19, and 20. Moreover our result does not impose any constraints on the statistical properties of the non-equilibrium interacting central region.

We have then applied our theoretical framework to a model system of electron-vibron interacting nanojunction. We have analyzed in detail the domain of validity of Landauer-like approaches, i.e., without renormalization of the contacts, to describe the conductance and the IETS of such a nonequilibrium many-body interacting system.

Our results confirm that generally Landauer-like approaches are not adequate to describe the transport properties of such interacting systems for the whole range of applied biases (linear to highly nonlinear regime) and for all the transport regimes (good, metallic-like to mediocre, insulator-like conductors). In general, the correct transport properties are only obtained from exact nonequilibrium many-body Green's function calculations. However, there exist a certain number of conditions in which a Landauer-like approach can reproduce fairly well either the conductance or the IETS signal. For example, renormalization of the Green's functions in a Landauer-like approach is already sufficient to account qualitatively for the inelastic features in the IETS signal for the resonant transport regime, i.e., I^{LL} gives features at ω_0 in the IETS signal. However this is not the case for the off-resonant transport regime, for which the Landauer-like cur-

rent I^{LL} fails to reproduce the inelastic features in the IETS signal.

Finally, we believe that higher-order diagrams, as studied in Ref. 49, may change the detailed features of the IETS signal, especially for higher energy excitations. However, the accuracy to/with which the Green's functions are calculated (Born approximation/Hartree-Fock or beyond) does not alter the main conclusions of our work concerning the applicability of Landauer-like current formula versus exact derivation of the current.

ACKNOWLEDGMENT

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APPENDIX A: RELATIONSHIPS BETWEEN GREEN'S FUNCTIONS

In order to keep the formalism simple, let us first consider that the Green's functions and the corresponding self-energies are simply complex functions of ω , i.e., we are dealing with an interacting central region containing only one site/one electronic level (SSSM model).

When the system is at equilibrium ($f_L=f_R=f^{\text{eq}}$), the lesser (greater) Green's function is related to the advanced and retarded Green's functions,

$$G^{<,\text{eq}}(\omega) = -f^{\text{eq}}(\omega) [G^{r,\text{eq}}(\omega) - G^{a,\text{eq}}(\omega)] \quad (\text{A1})$$

and

$$G^{>,\text{eq}}(\omega) = -[f^{\text{eq}}(\omega) - 1] [G^{r,\text{eq}}(\omega) - G^{a,\text{eq}}(\omega)]. \quad (\text{A2})$$

These relationships are at the center of the fluctuation-dissipation theorem for equilibrium. They can also be recast as follows:

$$G^{>,\text{eq}}(\omega) = -e^{(\omega-\mu_0)/kT} G^{<,\text{eq}}(\omega) \quad (\text{A3})$$

and they then define a relationship between the greater and lesser Green's functions for statistical averages in the grand canonical ensemble at finite temperature (the so-called Kubo-Martin-Schwinger boundary conditions^{35,54}). Similar relationships also exist for the self-energies $\Sigma^{<,>,r,a}$ (see, for example, Ref. 63).

For nonequilibrium conditions, there is no unique Fermi level at finite bias (or no unique temperature if $T_L \neq T_R$) in the whole system, and the relationships given above by Eqs. (A1)–(A3) no longer hold. This is an important feature of the nonequilibrium formalism for which conventional equilibrium statistics need to be reformulated.⁶⁴ However, the self-consistent calculations of the Green's functions and self-energies in the nonequilibrium case permit us to define new nonequilibrium distributions. For example, the nonequilibrium distribution $f^{\text{NE}}(\omega)$ is defined from the Green's functions as follows:

$$G^<(\omega) = -f^{\text{NE}}(\omega) [G^r(\omega) - G^a(\omega)]. \quad (\text{A4})$$

This definition is reminiscent of the so-called Kadanoff-Baym ansatz which has been generalized to the nonequilib-

rium conditions and to the time representation of the Green's functions (see, for example, Refs. 65 and 66). Similarly, we also define the nonequilibrium distribution $f_{\text{int}}^{\text{NE}}(\omega)$ from the interaction self-energies as follows:

$$\Sigma_{\text{int}}^{<}(\omega) = -f_{\text{int}}^{\text{NE}}(\omega)[\Sigma_{\text{int}}^r(\omega) - \Sigma_{\text{int}}^a(\omega)]. \quad (\text{A5})$$

There is no *a priori* reason for these two nonequilibrium distribution functions to be equal to each other at nonequilibrium. Both distribution functions contain information about both the nonequilibrium and the many-body interaction effects in the system^{64,67}. However, at equilibrium, these distribution functions are as expected equal to each other and to the conventional Fermi-Dirac equilibrium statistics $f^{\text{NE}} = f_{\text{int}}^{\text{NE}} = f^{\text{eq}}$.

As an example, the nonequilibrium distribution function $f_0^{\text{NE}}(\omega)$ for a noninteracting system⁶⁸ is given by the weighted averaged of $f_{L,R}$ by the coupling at each contact $\Gamma_{L,R}$,

$$f_0^{\text{NE}}(\omega) = \frac{f_L(\omega)\Gamma_L(\omega) + f_R(\omega)\Gamma_R(\omega)}{\Gamma_L(\omega) + \Gamma_R(\omega)}. \quad (\text{A6})$$

The asymptotic values of distribution functions are defined from behavior of the Green's functions and self-energies at large ω , and follow the conventional statistics,

$$f^{\text{NE}}(\omega) = f_{\text{int}}^{\text{NE}}(\omega) = f_0^{\text{NE}}(\omega) = f_\alpha(\omega) = f^{\text{eq}}(\omega) \begin{cases} = 1, & \omega \rightarrow -\infty \\ = 0, & \omega \rightarrow +\infty. \end{cases} \quad (\text{A7})$$

At equilibrium, one recovers the equilibrium statistics for any distribution function $f_0^{\text{NE}} = f^{\text{eq}}$.

Now, we can generalize our formalism to central regions containing several electronic states. The distribution functions then become matrices \mathbf{f} (Refs. 13, 45, and 69) with elements f_{nm} given by

$$X_{nm}^{<} = -\sum_l f_{nl}(X_{lm}^r - X_{lm}^a), \quad (\text{A8})$$

where X is either a Green's function G or a self-energy Σ , and the indices n,m are appropriate indices to label the electronic states of the central region.

APPENDIX B: THE Ng ANSATZ

Here again, we consider in the following mathematical developments that the Green's functions and the self-energies are simply complex functions. Extension to matrices is rather straightforward but must be done with care using the notations and definitions given in the main text and in Appendix A.

Using the definition of $f_{\text{int}}^{\text{NE}}(\omega)$,

$$\begin{aligned} \Sigma_{\text{int}}^{<}(\omega) &= -f_{\text{int}}^{\text{NE}}(\omega)[\Sigma_{\text{int}}^r(\omega) - \Sigma_{\text{int}}^a(\omega)] \\ &= -f_{\text{int}}^{\text{NE}}(\omega)[\Sigma_{\text{int}}^{>}(\omega) - \Sigma_{\text{int}}^{<}(\omega)], \end{aligned} \quad (\text{B1})$$

and the fact that $f_\alpha(\omega) = -i\Sigma_L^{<}(\omega)/\Gamma_L(\omega)$, we find after more formal manipulations that the renormalization functional $\Lambda(\omega)$ [Eq. (10)] can be re-expressed as

$$\Lambda(\omega) = 1 + i \frac{\Sigma_L^{<}(\omega)\Sigma_{\text{int}}^{>}(\omega) - \Sigma_L^{>}(\omega)\Sigma_{\text{int}}^{<}(\omega)}{\Sigma_L^{<}(\omega)\Gamma_R(\omega) - \Sigma_R^{<}(\omega)\Gamma_L(\omega)}. \quad (\text{B2})$$

After noticing that $\Sigma_L^{<}(\omega)\Gamma_R(\omega) - \Sigma_R^{<}(\omega)\Gamma_L(\omega) = i\Sigma_L^{<}(\omega)\Sigma_{L+R}^{>} - i\Sigma_L^{>}(\omega)\Sigma_{L+R}^{<}$, we can finally obtain a compact form for $\Lambda(\omega)$,

$$\Lambda(\omega) = \frac{\Sigma_L^{<}(\omega)\Sigma_{L+R}^{>}(\omega) - \Sigma_L^{>}(\omega)\Sigma_{L+R}^{<}(\omega)}{\Sigma_L^{<}(\omega)\Sigma_{L+R}^{>}(\omega) - \Sigma_L^{>}(\omega)\Sigma_{L+R}^{<}(\omega)}, \quad (\text{B3})$$

which is another way of expressing the important result of this paper given in Eq. (10).

Now we are going to relate our principal results to previous studies using the Ng ansatz.^{19–21} The Ng ansatz, developed to study the Anderson model out of equilibrium,^{19–21} is based on using an apparently more convenient way to express the full lesser (greater) self-energy in terms of the lesser (greater) self-energy for the noninteracting system,

$$\Sigma^{<,>}(\omega) = \Sigma_{L+R}^{<,>}(\omega)\bar{\Lambda}(\omega), \quad (\text{B4})$$

where $\Sigma^x = \Sigma_{L+R}^x + \Sigma_{\text{int}}^x$ and $\Sigma_{L+R}^x = \Sigma_L^x + \Sigma_R^x$, and $\bar{\Lambda}$ is a dynamical “renormalization” quantity, to be determined from the condition $\Sigma^> - \Sigma^< = \Sigma^r - \Sigma^a$. We will show below that this ansatz actually implies strong constraints on the statistics on the nonequilibrium interacting systems.

By using the Ng ansatz to express the lesser and greater self-energies $\Sigma^{<,>}(\omega)$ in Eq. (B3), one can easily see that our renormalization functional $\Lambda(\omega)$ given by Eq. (B3) is just equal to the dynamical quantity $\bar{\Lambda}(\omega)$ of the Ng ansatz: $\Lambda(\omega) = \bar{\Lambda}(\omega)$. We then recover all the expressions for the current previously derived in Refs. 16, 17, and 19–21 from our main results Eqs. (8)–(10).

However, the Ng ansatz presents some intrinsic limitations. To prove this, it is sufficient to calculate the nonequilibrium distribution function $f_{\text{int}}^{\text{NE}}$ within the Ng ansatz. Starting from the definition of $f_{\text{int}}^{\text{NE}}(\omega)$, i.e.,

$$f_{\text{int}}^{\text{NE}}(\omega) = -\Sigma_{\text{int}}^{<}(\omega)/[\Sigma_{\text{int}}^r(\omega) - \Sigma_{\text{int}}^a(\omega)], \quad (\text{B5})$$

it is straightforward to show that $f_{\text{int}}^{\text{NE}}$ is then given by

$$\begin{aligned} f_{\text{int}}^{\text{NE}}(\omega) &= -\frac{\Sigma_{L+R}^{<}(\omega)(\bar{\Lambda}(\omega) - 1)}{(\Sigma_{L+R}^{>} - \Sigma_{L+R}^{<})(\bar{\Lambda} - 1)} \\ &= \frac{f_L(\omega)\Gamma_L(\omega) + f_R(\omega)\Gamma_R(\omega)}{\Gamma_L(\omega) + \Gamma_R(\omega)} = f_0^{\text{NE}}(\omega), \end{aligned} \quad (\text{B6})$$

the nonequilibrium distribution function for the noninteracting system.

Just to confirm the consistency of our derivations, if we use the above result $f_{\text{int}}^{\text{NE}}(\omega) = f_0^{\text{NE}}(\omega)$ in the definition of our renormalization functional $\Lambda(\omega)$ given by Eq. (10) and the Ng ansatz for $\Sigma_{\text{int}}^{<,>}$, i.e., $\Sigma_{\text{int}}^{<,>}(\omega) = \Sigma_{L+R}^{<,>}(\omega)(\bar{\Lambda}(\omega) - 1)$, we find again and consistently that $\Lambda(\omega) = 1 + [\bar{\Lambda}(\omega) - 1] = \bar{\Lambda}(\omega)$, as expected.

However we have found⁶⁷ that the condition $f_{\text{int}}^{\text{NE}}(\omega) = f_0^{\text{NE}}(\omega)$ implies necessarily that $f^{\text{NE}}(\omega) = f_0^{\text{NE}}(\omega)$. In other terms, the Ng ansatz implies that the full nonequilibrium distribution f^{NE} of the interacting system, as well as $f_{\text{int}}^{\text{NE}}$, are equal to the nonequilibrium distribution function for the non-

interacting system. This is a condition that is in contradiction with the fact that both distribution functions should simultaneously include both the nonequilibrium effects and the many-body interaction effects. In fact $f^{\text{NE}}(\omega)$ is a functional of both nonequilibrium distribution functions of the noninteracting system $f_0^{\text{NE}}(\omega)$ and of the many-body interaction $f_{\text{int}}^{\text{NE}}(\omega)$.^{64,67}

Hence we conclude that our expression for the renormalization of the coupling at the contact $\Lambda(\omega)$ given by Eq. (10) is more general than the definition used in the Ng ansatz. The latter is not taking fully into account the interaction effects at nonequilibrium. It actually corresponds to a lowest-order expansion of the full nonequilibrium distribution in terms of only the nonequilibrium distribution function of the noninteracting system⁶⁷ or in other words, the Ng ansatz considers that the statistics of the interacting central region is dominated by that of the noninteracting leads at nonequilibrium,

and that the interaction effects in the central region do not affect its nonequilibrium statistics.

Finally we would like to mention that it is however possible to recover the Ng ansatz from our results in the limit of low-energy scales,⁶⁷ i.e., when $(\omega - \mu_\alpha) \sim 0$ and then $\exp(\omega - \mu_\alpha)/kT \sim 1$. This implies that although approximate, the Ng ansatz might be good enough to describe low-energy excitations like, for example, the Kondo effect in correlated electron systems, which gives a sharp feature in the spectral density around the Fermi level at equilibrium or split Kondo peaks around the leads' Fermi levels at nonequilibrium.⁷⁰ However, such an ansatz will most probably fail to describe systems for which the interaction (electron-phonon, electron-plasmon) is restricted on an energy scale defined by the phonon (plasmon) frequency, which is finite and not necessarily small.^{49,67}

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