

# Nonequilibrium distribution functions for quantum transport: Universality and approximation for the steady state regime

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We derive a general expression for the electron nonequilibrium (NE) distribution function in the context of steady state quantum transport through a two-terminal nanodevice with interactions. The central idea for the use of NE distributions for open quantum systems is that both the NE and many-body (MB) effects are taken into account in the statistics of the finite size system connected to reservoirs. We develop an alternative scheme to calculate the NE steady state properties of such systems. The method, using NE distribution and spectral functions, presents several advantages, and is equivalent to conventional steady state NE Green's function (NEGF) calculations when the same level of approximation for the MB interaction is used. The advantages of our method resides in the fact that the NE distribution and spectral functions have better analytic behavior for numerical calculations. Furthermore, our approach offers the possibility of introducing further approximations, not only at the level of the MB interaction as in NEGF, but also at the level of the functional form used for the NE distributions. For the single-level model with electron-phonon coupling we have considered, such approximations provide a good representation of the exact results, for either the NE distributions themselves or the transport properties. We also derive the formal extensions of our method for systems consisting of several electronic levels and several vibration modes.

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## I. INTRODUCTION

The understanding of irreversible phenomena including the nonequilibrium (NE) steady state is a long-standing problem of quantum statistical mechanics. With recent experimental developments, it is now possible to measure the transport properties through nanoscale systems. These can be either the electronic charge transport or heat transport. Both properties, i.e., thermoelectric transport, have recently been measured simultaneously [1]. Such properties exhibit many important new features in comparison with conduction through macroscopic systems. In particular, the interactions, such as the Coulomb interaction between electrons and scattering from atomic vibrations, become critically important in nanoscale objects, especially in single organic molecules [2,3].

Modeling such transport properties is still a challenge since one needs to be able to describe the system at the atomic level in a realistic manner, and one needs to use a formalism for the quantum transport that takes full account of the NE conditions (full nonlinear response) and the many-body (MB) interaction.

The nonequilibrium Green's function (NEGF) seems to be, at the present moment, the best way to tackle the problem. However, NEGF calculations for realistic systems are difficult to achieve, beyond mean-field-like approximations or quasiequilibrium regimes, since the calculations of the MB effects for a large number of electronic (and vibronic) degrees of freedom are extremely demanding. Alternatively, the density-functional (DF)-based theories can handle large systems, but unfortunately treat the interaction (between electrons, for example) on a mean-field-like basis and the

corresponding functionals are not necessarily optimized, or even valid, for the NE conditions.

In this paper, we present an alternative approach based on the use of NE distribution and spectral functions. On the one hand, such an approach is, in principle, strictly equivalent to the steady state NEGF technique, since there is a one-to-one equivalence between the Green's functions (GFs) and the NE distribution and spectral functions. On the other hand, approximations for the MB effects (in the presence of NE conditions) seem to be more easily introduced in the NE distribution, while keeping a clear physical interpretation. Furthermore, the use of approximated NE distributions may offer an alternative approach for future implementations in DF-based calculations for large systems.

In earlier studies, we have already started developing and using the concept of NE distribution functions. This was done in a critical analysis of the applicability of the Landauer formalism for NE current in the presence of interactions [4], and in the study of the NE charge susceptibility and its relation with the nonlinear dynamical conductance [5].

In this paper, we develop in detail our approach using NE distribution and spectral functions, and provide numerical applications. The paper is organized as follows. In Sec. II, we define the general steady state transport setup. We start by considering a model system in Sec. III and provide all the analytical results for the NE distributions. Section III A concerns the general properties of the NE distributions. In Sec. III B, we develop an algorithm for performing NE calculations. Numerical applications are provided in Sec. III C, where we show examples of the NE distributions for a model of an electron-phonon interacting system. The performances of the exact and approximated NE distributions are studied in this section. The generalization of our approach to more realistic systems is provided in Sec. IV. Finally, we comment on our results and conclude our study in Sec. V.

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## II. STEADY STATE QUANTUM TRANSPORT

We consider a system consisting of a central region  $C$  connected to two noninteracting Fermi seas. The left ( $L$ ) and right ( $R$ ) electrodes are at their own equilibrium, with a Fermi distribution  $f_\alpha(\omega)$  defined by their respective chemical potentials  $\mu_\alpha$  and temperatures  $T_\alpha$  ( $\alpha = L, R$ ). The central region  $C$  connected to the leads contains an interaction characterized by a self-energy  $\Sigma_{\text{int}}(\omega)$  in the NEGF formalism. Furthermore, the specific model used for the leads does not need to be specified at the moment, as long as the leads can also be described by an embedding self-energy  $\Sigma_\alpha(\omega)$  in the electron GF of the central region.

The possibility of reaching a steady state regime in such a two-terminal device has been explored by many authors. The full time-dependent NEGF formalism and the influence of bound states in the central region have been studied in Refs. [6–10]. Rigorous mathematical methods based on the  $C^*$  algebra have been used to study the existence and stability of such a NE steady state, i.e., its independence of the way the division into subsystems and reservoirs is performed and its stability against local perturbations, in the absence [11–14] and in the presence of an interaction [15,16].

For an established steady state regime, it is expected that some formal advantages may be given by an approach to NE processes in which the Gibbs-like ensembles play a prominent role. The construction of such Gibbs-like ensembles for the NE steady state can be obtained either by using the McLennan-Zubarev approaches [12,17–23] or the NE density matrix approach developed by Hershfield in Ref. [24]. The latter has been extensively used for calculating quantum electron transport properties, with or without an interaction [25–33].

In the following, we show that the NE statistics of the open quantum system, i.e., the central region  $C$ , contains information not only about the NE conditions but also about the MB interaction.

## III. THE SINGLE-IMPURITY MODEL

We now consider a model for the central region made of a single electron level in the presence of an interaction. In this section all quantities are either real or complex number functions of a single energy argument.

### A. The NE distribution $f^{\text{NE}}$

In a recent paper [34], we have shown, using McLennan-Zubarev and Hershfield approaches, that the steady state can be interpreted as an effective equilibrium state with a corresponding NE density matrix, or equivalently, with a corresponding NE statistics.

Such a NE statistic can be defined by a NE distribution function  $f^{\text{NE}}(\omega)$ . It enters the relation between the different GFs defined in the central region  $C$  as follows:

$$G_C^{\lessgtr}(\omega) = -f^{\text{NE},\lessgtr}(\omega)(G_C^r(\omega) - G_C^a(\omega)), \quad (1)$$

with  $f^{\text{NE},<}(\omega) = f^{\text{NE}}$  and with  $f^{\text{NE},>}(\omega) = f^{\text{NE}} - 1$ . We recall that the spectral function  $A_C(\omega)$  of the central region is obtained from  $A_C(\omega) = (G_C^a - G_C^r)/i2\pi$ . Equation (1) bears resemblance to the so-called Kadanoff-Baym ansatz [35,36],

but as we have shown in Ref. [34], it is a strictly exact result for the steady state regime.

At equilibrium,  $f^{\text{NE}}$  is simply the Fermi distribution  $f^{\text{eq}}$ . Out of equilibrium, the distribution function will depend on the setup, i.e., on the forces driving the system (gradient of chemical potential and/or temperature between the leads), and on the interaction present in the region  $C$ .

In the absence of an interaction, the NE distribution function for the electron is simply given by [26,37]

$$f_0^{\text{NE}}(\omega) = \frac{\Gamma_L(\omega)f_L(\omega) + \Gamma_R(\omega)f_R(\omega)}{\Gamma_L(\omega) + \Gamma_R(\omega)}, \quad (2)$$

where  $\Gamma_\alpha(\omega) = i(\Sigma_\alpha^> - \Sigma_\alpha^<)(\omega)$  is the spectral function of the embedding (lead  $\alpha$ ) self-energy. It is simply a double-step function, with more or less steep steps (depending on the temperature  $T_L$  and  $T_R$ ) located around  $\omega = \mu_L$  and  $\omega = \mu_R$ , and separated by  $\mu_L - \mu_R = eV$  ( $V$  is the applied bias).

In the presence of an interaction in the central region  $C$ , the NE distribution is given by [34]

$$\begin{aligned} f^{\text{NE}}(\omega) &= \frac{G_C^<}{G_C^a - G_C^r} = \frac{G_C^r \Sigma^< G_C^a}{G_C^r ((G_C^r)^{-1} - (G_C^a)^{-1}) G_C^a} \\ &= \frac{\Sigma_L^< + \Sigma_R^< + \Sigma_{\text{int}}^<}{\Sigma^a - \Sigma^r}. \end{aligned} \quad (3)$$

Using the definitions  $\Sigma_L^< + \Sigma_R^< = i\Gamma_{L+R}f_0^{\text{NE}}$  and  $\Sigma^a - \Sigma^r = -(\Sigma^> - \Sigma^<) = i\Gamma_{L+R} - (\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<)$ , with  $\Gamma_{L+R}(\omega) = \Gamma_L(\omega) + \Gamma_R(\omega)$ , we obtain

$$f^{\text{NE}}(\omega) = \frac{f_0^{\text{NE}}(\omega) - i\Sigma_{\text{int}}^<(\omega)/\Gamma_{L+R}(\omega)}{1 + i(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<)/\Gamma_{L+R}}. \quad (4)$$

Equation (4) is the ‘‘universal’’ expression of the electron NE distribution function. It is universal with respect to the interaction, in the same sense that the GFs have a universal expression via the use of the interaction self-energies. However, as expected for NE conditions, the NE distribution function is not as universal as its equilibrium counterpart, since it depends on both the setup that drives the system out of equilibrium (via  $f_0^{\text{NE}}$ ) and on the MB interaction  $\Sigma_{\text{int}}^{\lessgtr}$  (which are themselves dependent on the NE conditions). We comment more on these properties in Appendix A.

From Eq. (4), we can see that the NE distribution  $f^{\text{NE}}$  arises from two terms,

$$f^{\text{NE}}(\omega) = \tilde{f}_0^{\text{NE}}(\omega) + \delta f^{\text{NE}}(\omega), \quad (5)$$

a dynamically renormalized distribution  $\tilde{f}_0^{\text{NE}} = f_0^{\text{NE}}(\omega)/\mathcal{N}(\omega)$ , with the renormalization  $\mathcal{N}(\omega)$  given by the sum of the spectral functions of the leads  $\Gamma_{L+R} = \sum_{\alpha=L,R} i(\Sigma_\alpha^> - \Sigma_\alpha^<)$  and of the interaction  $\Gamma_{\text{int}} = i(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<)$ , and an extra term  $\delta f^{\text{NE}}$  corresponding to the inelastic processes given by  $\Sigma_{\text{int}}^<$ , and renormalized by the same factor  $\mathcal{N}(\omega)$ .

The noninteracting distribution  $f_0^{\text{NE}}$  is formed by two Fermi-Dirac distributions shifted by the bias  $V$ . However, the full NE distribution presents richer feature (peaks and dips) characteristics of the electron population redistribution arising from both the NE and interaction effects. One can obtain both accumulation or depletion (i.e., population inversion) in some energy windows, and such features in the

NE distribution provide information about the efficiency of relaxation/equilibration processes in the system.

Furthermore, another important property of the NE distribution  $f^{\text{NE}}$  is related to its functional form. Indeed, any Feynmann diagrams for the interaction self-energy  $\Sigma_{\text{int}}$  (taken at any order and for electron-electron  $e$ - $e$  or electron-phonon  $e$ -ph interactions) is expressed in terms of the different electron GFs and phonon GFs. The renormalization of the phonon GFs, if present, is also obtained from another set of diagrams using the electron GFs (in the case of  $e$ -ph interactions).

Since all GFs [either the retarded, the advanced, or the lesser, the greater, or the (anti)time order] can be expressed in terms of spectral function  $A_C$  alone or in terms of both the spectral function and the NE distribution [see Eq. (1)], any self-energy is a functional of the spectral functions and of the NE distribution function. In Appendix C, we show explicitly how such a functional dependence is obtained by considering different lowest-order diagrams for the self-energies in case of both  $e$ - $e$  and  $e$ -ph interactions.

Therefore, from the general expression Eq. (4) defining  $f^{\text{NE}}$ , we can conclude that  $f^{\text{NE}} = f^{\text{NE}}[f^{\text{NE}}(\omega), A_C(\omega)]$ . The fact that  $f^{\text{NE}}$  is a functional of itself and of the spectral function permits us to devise an approach to solve self-consistently the problem by using an iterative scheme. Such a scheme is developed in the next section and bears resemblance to conventional self-consistent NEGF calculations.

## B. Algorithm for NE calculations

The method we present in this section has, however, some advantages compared to conventional NEGF calculations. First of all, we are now dealing with two real functions  $f^{\text{NE}}(\omega)$  and  $A_C(\omega)$  instead of complex number functions for the GFs. More importantly, these two functions have well behaved (for numerical purposes) asymptotic limits: The spectral function  $A_C(\omega)$  has a finite energy support, i.e.,  $A_C(\omega) \neq 0$  for  $\omega \in [\omega_{\min}, \omega_{\max}]$  otherwise  $A(\omega) = 0$ , and  $f^{\text{NE}}(\omega) = 1$  for  $\omega < D_{\omega}^{\text{NE}}$  and  $f^{\text{NE}}(\omega) = 0$  for  $\omega > D_{\omega}^{\text{NE}}$  where the energy domain  $D_{\omega}^{\text{NE}}$  is roughly the bias window  $D_{\omega}^{\text{NE}} = [\min(\mu_L, \mu_R), \max(\mu_L, \mu_R)] \pm \text{several } kT$ .

Hence by using only  $f^{\text{NE}}(\omega)$  and  $A_C(\omega)$ , we avoid having to deal with the slow decaying behavior in  $1/\omega$  of the real part of the advanced and retarded GFs and self-energies. Such a slow decay in  $1/\omega$  comes from the Fourier transform of the Heaviside function defining the causality in the retarded (the anticausality in the advanced) quantities. We are not obliged to work with large (i.e., long-ranged) energy grids. In principle, one should work with a grid larger than  $D_{\omega}^{\text{NE}}$  in order to include the possible effects of “hot electrons” excited well above the bias window due to the interaction. In practice, we have found that the energy grid could be only the support of the spectral function  $[\omega_{\min}, \omega_{\max}]$ .

### 1. The algorithm

The algorithm to perform NE steady state calculations is as follows:

(1) Start with an initial ( $n = 0$ ) spectral function  $A^{(n)}(\omega)$  and NE distribution  $f^{\text{NE}(n)}(\omega)$ , for example, those corresponding to the noninteracting case:  $A^{(0)}(\omega) = -\text{Im } mG_0^r(\omega)/\pi$  and  $f^{\text{NE}(0)}(\omega) = f_0^{\text{NE}}(\omega)$ .

(2) Calculate the corresponding initial self-energies  $\Sigma_{\text{int}}^{\leq(n)}$  for the chosen model of MB interactions.

(3) Calculate the next iteration NE distribution  $f^{\text{NE}(n+1)}(\omega)$  from Eq. (4) as follows:

$$f^{\text{NE}(n+1)} = \frac{f_0^{\text{NE}} \Gamma_{L+R} - i \Sigma_{\text{int}}^{\leq(n)}}{\Gamma_{L+R} + i (\Sigma_{\text{int}}^{\geq(n)} - \Sigma_{\text{int}}^{\leq(n)})}, \quad (6)$$

with  $\Sigma_{\text{int}}^{\leq(n)} = \Sigma_{\text{int}}^{\leq} [f^{\text{NE}(n)}, A_C^{(n)}]$ . Note that the quantities  $i \Sigma_{\text{int}}^{\leq(n)}$  are also real functions.

(4) Calculate the next iteration spectral function from either *method (a)*, by using the following expression,

$$A_C^{(n+1)}(\omega) = \frac{f^{\text{NE}(n)}(\omega)}{f^{\text{NE}(n+1)}(\omega)} A_C^{(n)}(\omega) \quad (7)$$

or *method (b)*, by using the definition of the spectral functions  $i2\pi A_C(\omega) = G_C^a(\omega) - G_C^r(\omega) = G_C^< - G_C^> = G_C^r(\Sigma^< - \Sigma^>)G_C^a$ , by which we define

$$2\pi A_C^{(n+1)}(\omega) = \mathcal{G}^{r(n)}(\Gamma_{L+R} + i(\hat{\Sigma}_{\text{int}}^{\geq(n)} - \hat{\Sigma}_{\text{int}}^{\leq(n)}))\mathcal{G}^{a(n)}, \quad (8)$$

where  $\mathcal{G}^{r/a(n)}$  should be considered as intermediate (dummy) functions defined from the  $n$ th iteration spectral function  $A_C^{(n)}(\omega)$  as

$$\mathcal{G}^{r/a(n)}(\omega) = \mathcal{H}[\pi A_C^{(n)}(\omega)] \mp i\pi A_C^{(n)}(\omega), \quad (9)$$

where  $\mathcal{H}[f(x)]$  is the Hilbert transform of function  $f(x)$ , i.e.,  $\mathcal{H}[f(x)] = 1/\pi \text{P} \int dy f(y)/(x - y)$  and  $\hat{\Sigma}_{\text{int}}^{\leq(n)}$  is an intermediate updated version of the self-energy obtained from  $\hat{\Sigma}_{\text{int}}^{\leq(n)} = \Sigma_{\text{int}}^{\leq} [f^{\text{NE}(n+1)}, A_C^{(n)}]$ .

(5) Ensure normalization of  $A_C^{(n+1)}$  when using approximated functionals for the NE distribution such as  $f_{\text{LOE}}^{\text{NE}}$  or  $f_{(1)}^{\text{NE}}$  (see below).

(6) Repeat the iteration process, from step (3), until the desired convergence is achieved [either for the NE distribution  $f^{\text{NE}(n+1)}$  or for the spectral function  $A^{(n+1)}(\omega)$  or for both].

It should be noted that, similarly to the spectral functions, the spectral “densities” of the self-energy of the leads  $\Gamma_{L+R}(\omega)$ , and of the interaction self-energy  $i(\Sigma_{\text{int}}^{\geq} - \Sigma_{\text{int}}^{\leq})$  are bounded, i.e., they have zero values outside an energy interval which is roughly the same as  $[\omega_{\min}, \omega_{\max}]$ . Hence we do not have to worry about the long-ranged dependence in  $1/\omega$  of the real part of  $\mathcal{G}^{r/a(n)}$ , and we recover spectral functions  $A^{(n+1)}(\omega)$  which exist only on a finite energy support.

Furthermore, method (a) for the calculation of the spectral function presents the great advantage of being extremely simple, in comparison to method (b) [38]. However, we have noticed that, in some cases when the initial spectral function of the iterative loop is too different from the expected result, the convergence process might be slower (if not possible at all) for method (a) than for method (b). Hence method (b) appears to be more robust upon the choice of the initial conditions. It is entirely possible to combine both schemes in the same algorithm, starting first with method (b), and when some degree of convergence is reached, switching to method (a) to obtain a more accurate level of convergence.

## 2. Approximated NE distributions

The method devised in the previous section can appear at first glance as just another reformulation of conventional NEGF calculations. One performs calculations with another set of two independent (but interrelated) functions: the NE distribution  $f^{\text{NE}}$  and the spectral function  $A_C$ . In the conventional NEGF technique, one deals instead with the two independent GFs,  $G_C^>$  and  $G_C^<$ . As mentioned above, there is indeed a one-to-one correspondence between these two sets of functions.

However, our method offers many advantages and not only on the numerical point of view, as explained previously. Indeed, as the NE distribution is a functional of itself and of the spectral function, it offers a more direct and natural way of performing approximated calculations, by considering some specific subsets of inelastic processes. Such approximations are advantageous to minimize the computational cost of the calculations, which is an important point for future applications to large and more realistic systems.

Ultimately  $f^{\text{NE}}$ , given by Eq. (4), can be expressed as an infinite series expansion in terms of the noninteracting NE distribution  $f_0^{\text{NE}}$ , the spectral function  $A_C$ , and the interaction parameters ( $\gamma_0$  or  $v_q$ ). Therefore, instead of performing the calculations with the exact expression Eq. (4), we can always truncate the series expansion to a desired level of accuracy (i.e., selecting a specific subset of inelastic processes), suitable for the system and the properties under study.

We provide, in the next section, some examples of approximated NE distributions and compare their performance against exact calculations using the full  $f^{\text{NE}}$  distribution. We recall that the latter are strictly equivalent to NEGF calculations (with the same model of self-energies).

## C. Numerical application

We now consider numerical applications for a model of an  $e$ -ph interacting system, and we test the different approximations available for the functional forms of the NE distribution.

### 1. Model of electron-phonon interacting system

The Hamiltonian for the region  $C$  is

$$H_C = \varepsilon_0 d^\dagger d + \hbar\omega_0 a^\dagger a + \gamma_0 (a^\dagger + a) d^\dagger d, \quad (10)$$

where  $d^\dagger$  ( $d$ ) is the creation (annihilation) operator of an electron in the molecular level  $\varepsilon_0$ . The electron is coupled, via its charge density  $d^\dagger d$ , to the vibration mode (phonon) of energy  $\omega_0$  and the strength of the coupling is given by the constant  $\gamma_0$ , and  $a^\dagger$  ( $a$ ) creates (annihilates) a quantum in the vibron mode  $\omega_0$ .

For the transport setup, the central region  $C$  is connected to two ( $L$  and  $R$ ) one-dimensional semi-infinite tight-binding chains via the hopping integrals  $t_{0L}$  and  $t_{0R}$ . The corresponding  $\alpha = L, R$  self-energy is obtained from the GF at the end of the semi-infinite tight-binding chain and is given by  $\Sigma'_\alpha(\omega) = t_{0\alpha}^2 e^{ik_\alpha(\omega)} / \beta_\alpha$ . A dispersion relation links the energy  $\omega$  with the momentum  $k_\alpha$  of an electron in the lead  $\alpha$ :  $\omega = \varepsilon_\alpha + 2\beta_\alpha \cos k_\alpha(\omega)$ . The parameters  $\varepsilon_\alpha$  and  $\beta_\alpha$  are the on-site and off-diagonal elements of the tight-binding chains. With such a

choice of lead self-energy, we go beyond the wideband limit (unless  $\beta_\alpha$  is much larger than any other parameters).

The self-energies  $\Sigma_{\text{int}}$  for the interaction between the electron and the vibration mode are calculated using the Born approximation [39,40]. Their expressions are given in Appendix B.

Finally, in the most general cases, the left and right contacts are different ( $\Gamma_L \neq \Gamma_R$ ) and there are asymmetric potential drops, i.e.,  $\mu_\alpha = \mu^{\text{eq}} + \eta_\alpha V$ , with the condition  $\Delta\mu = \mu_L - \mu_R = V$  (i.e.,  $\eta_L - \eta_R = 1$ ).

## 2. Examples of NE distributions

We provide typical examples of the charge redistribution in the central region induced by both the NE effects and the interaction. For a given model of interaction self-energies, the full self-consistent calculations provided by the algorithm in Sec. III B 1 are strictly equivalent to conventional NEGF calculations. Hence the results obtained for  $f^{\text{NE}}$  with our method (and full self-consistency) are equal to those obtained from NEGF-self-consistent Born approximation (SCBA) calculations [39,40].

Figure 1 shows how the NE distribution evolves upon increasing the NE conditions, i.e., the applied bias, for a typical set of parameters characterizing the off-resonant transport regime. One can clearly observe the difference between the noninteracting NE distribution  $f_0^{\text{NE}}$  and the full distribution  $f^{\text{NE}}$ . The latter presents features (peaks and dips) which correspond to the accumulation or depletion of the electron population induced by inelastic scattering effects. Such features are directly related to the peaks in the spectral function. This single example confirms explicitly that, generally,  $f^{\text{NE}} \neq f_0^{\text{NE}}$ , as shown analytically in Appendix A.

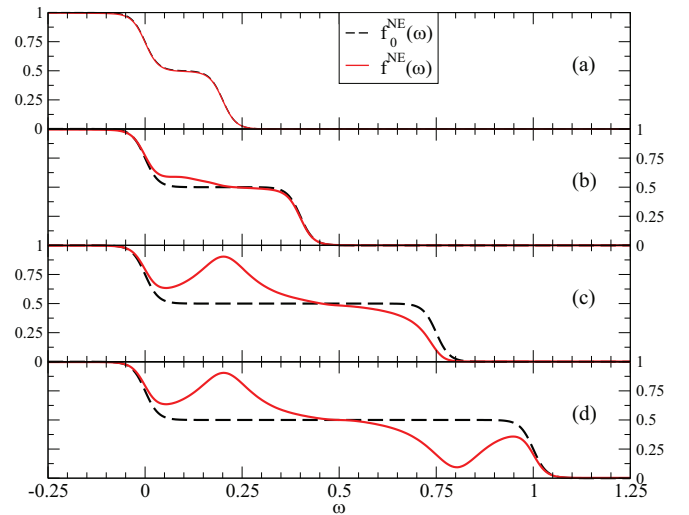


FIG. 1. (Color online) NE distribution functions for the off-resonant regime ( $\varepsilon_0 = 0.50$ ) and for different biases  $V$ . (a)  $V = 0.2 < \omega_0$ , (b)  $V = 0.4 \sim \omega_0$ , (c)  $V = 0.75 > \omega_0$ , (d)  $V = 1.0 \gg \omega_0$ . The NE distribution  $f^{\text{NE}}$  is completely different from the noninteraction NE distribution  $f_0^{\text{NE}}$  when  $V \geq \omega_0$ ; in this case inelastic processes occur and induce a redistribution of the electron population in the region  $C$ . The other parameters are  $\gamma_0 = 0.09$ ,  $\omega_0 = 0.3$ ,  $t_{0\alpha} = 0.15$ ,  $T_\alpha = 0.017$ ,  $\eta_L = 1$ ,  $\varepsilon_\alpha = 0$ ,  $\beta_\alpha = 2$ .

### 3. Approximated NE distributions

As mentioned in Sec. III B 2, for a given choice of interaction self-energies, our approach is fully equivalent to NEGF calculations. Both methods corresponds to a partial resummation of a family of diagrams associated with the interaction self-energy. However, we can further approximate the expression of the NE distribution Eq. (4). This corresponds to another way of partially resumming the diagrams corresponding to  $\Sigma_{\text{int}}$ .

A lowest-order expansion (LOE), in terms of the characteristic interaction parameter, gives an approximated NE distribution in the following form:

$$\begin{aligned} f^{\text{NE}}(\omega) &\sim \left( f_0^{\text{NE}}(\omega) - \frac{i \Sigma_{\text{int}}^<}{\Gamma_{L+R}} \right) \left( 1 - \frac{i(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<)}{\Gamma_{L+R}} \right) \\ &\sim f_0^{\text{NE}} \left( 1 - i \frac{\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<}{\Gamma_{L+R}} \right) - \frac{i \Sigma_{\text{int}}^<}{\Gamma_{L+R}} + O(\gamma_0^{n \geq 4}). \end{aligned} \quad (11)$$

Using the expressions for the self-energies  $\Sigma_{\text{int}}^{\gtrless}$  given in Appendix B for the limit  $N_{\text{ph}} = 0$ , we find that

$$\begin{aligned} f_{\text{LOE}}^{\text{NE}}(\omega) &= f_0^{\text{NE}}(\omega) + \frac{2\pi\gamma_0^2}{\Gamma_{L+R}} \{ A_C(\omega + \omega_0) f_0^{\text{NE}}(\omega + \omega_0) \\ &\quad \times [1 - f_0^{\text{NE}}(\omega)] - A_C(\omega - \omega_0) \\ &\quad \times [1 - f_0^{\text{NE}}(\omega - \omega_0)] f_0^{\text{NE}}(\omega) \}, \end{aligned} \quad (12)$$

where the terms in  $\gamma_0^2$  represent NE inelastic correction terms (to the noninteracting distribution  $f_0^{\text{NE}}$ ) arising from phonon emission by the electron and hole. Such correction terms are proportional to the ratio  $\gamma_0^2/\Gamma_{L+R}$ , where  $\Gamma_{L+R}$  represents the total escape (injection) rate of the electron or hole from (into) the central region C. The order of the interaction parameter is  $\gamma_0^2$  as in lowest-order perturbation theory.

Equation (12) represents the simplest functional form of the NE distribution  $f^{\text{NE}}(\omega) = f^{\text{NE}}[f_0^{\text{NE}}, A_C](\omega)$ . However, it is a lowest-order series expansion in terms of the parameter  $\gamma_0$  and is only valid for weak coupling, as we will show below. It should be noted that the inelastic processes can only occur when the bias  $V$  is larger than or equal to the excitation energy,  $V \geq \omega_0$ , otherwise the factors associated with phonon emission by the electron [ $f_0^{\text{NE}}(\omega + \omega_0)(1 - f_0^{\text{NE}}(\omega))$ ] or by the hole [ $f_0^{\text{NE}}(\omega)(1 - f_0^{\text{NE}}(\omega - \omega_0))$ ] are zero over the whole energy range [41].

Another possible approximation is to consider Eq. (4) using only the noninteracting distribution  $f_0^{\text{NE}}$  in the evaluation of the self-energies  $\Sigma_{\text{int}}^{\gtrless}$ . One then gets

$$\begin{aligned} f_{(1)}^{\text{NE}}(\omega) &= [f_0^{\text{NE}}(\omega)\Gamma_{L+R}(\omega) + 2\pi\gamma_0^2 A_C(\omega + \omega_0) \\ &\quad \times f_0^{\text{NE}}(\omega + \omega_0)] / \mathcal{N}(\omega), \end{aligned} \quad (13)$$

with  $\mathcal{N}(\omega) = \Gamma_{L+R}(\omega) + \Gamma_{\text{int}}(\omega)$  and

$$\begin{aligned} \Gamma_{\text{int}}(\omega) &= 2\pi\gamma_0^2 \{ A_C(\omega - \omega_0) [1 - f_0^{\text{NE}}(\omega - \omega_0)] \\ &\quad + A_C(\omega + \omega_0) f_0^{\text{NE}}(\omega + \omega_0) \}. \end{aligned} \quad (14)$$

In Fig. 2, we show different NE distributions calculated with different approximations: the noninteraction distribution  $f_0^{\text{NE}}$ ,

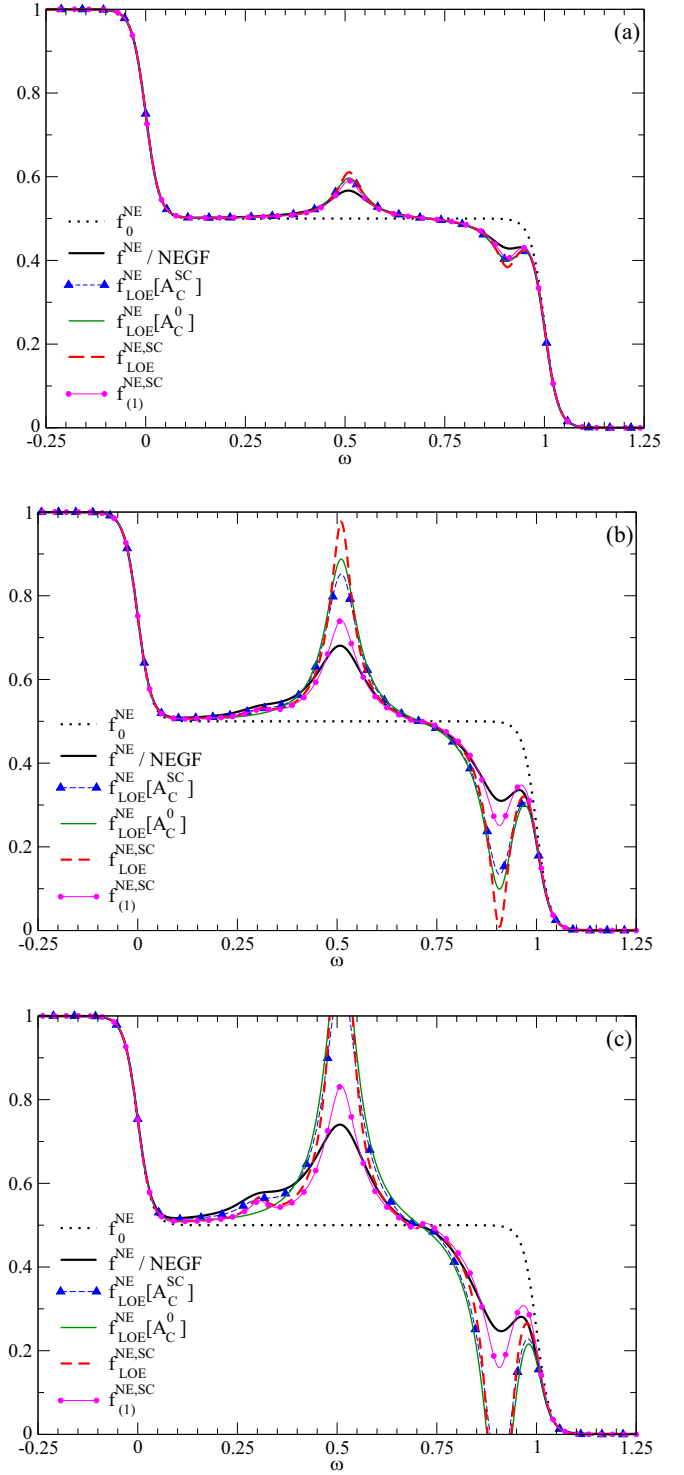


FIG. 2. (Color online) NE distribution functions for the off-resonant regime ( $\varepsilon_0 = 0.70$ ) for different approximations and for different coupling strengths  $\gamma_0$ . (a)  $\gamma_0 = 0.03$  ( $\gamma_0/\omega_0 = 0.15$ ), (b)  $\gamma_0 = 0.06$  ( $\gamma_0/\omega_0 = 0.3$ ), (c)  $\gamma_0 = 0.08$  ( $\gamma_0/\omega_0 = 0.4$ ). Only for weak coupling, all approximated NE distributions provide a good representation of the exact distribution  $f^{\text{NE}}$ . We recall that for fully self-consistent calculations, the results obtained for  $f^{\text{NE}}$  with our method are strictly equivalent to those obtained from NEGF calculations. See text for more detailed comments. The other parameters are  $V = 1.0$ ,  $\omega_0 = 0.2$ ,  $t_{0\alpha} = 0.22$ ,  $T_\alpha = 0.017$ ,  $\eta_L = 1$ ,  $\varepsilon_\alpha = 0$ ,  $\beta_\alpha = 2$ .

the full self-consistent distribution  $f^{\text{NE}}$ , and different approximations for the LOE distribution  $f_{\text{LOE}}^{\text{NE}}$  and  $f_{(1)}^{\text{NE}} \cdot f_{\text{LOE}}^{\text{NE}}[A_C^{\text{SC}}]$  is calculated from Eq. (12) using the full self-consistent spectral function  $A_C^{\text{SC}}(\omega)$ , and  $f_{\text{LOE}}^{\text{NE}}[A_C^0]$  is calculated from Eq. (12) using the noninteracting spectral function  $A_C^0(\omega)$ . Finally,  $f_x^{\text{NE,SC}}$  is obtained from a self-consistent calculation for the spectral function (see Sec. III B 1) using the functional form Eq. (12) for  $x = \text{LOE}$ , or Eq. (13) for  $x = (1)$ .

One can see in Fig. 2(a) that, for weak  $e$ -ph coupling, any approximations for  $f_{\text{LOE}}^{\text{NE}}$  give the same results, and provide a good representation of the exact  $f^{\text{NE}}$ . The distribution  $f_{(1)}^{\text{NE,SC}}$  provides a better representation for  $f^{\text{NE}}$ . The amplitude of  $f_{\text{LOE}}^{\text{NE}}$  is slightly different from  $f^{\text{NE}}$ , because it is obtained from a series expansion and is not fully renormalized by the factor  $\mathcal{N}(\omega)$ . Such a renormalization is, however, included in  $f_{(1)}^{\text{NE,SC}}$ .

For larger  $e$ -ph coupling, the difference between  $f_{\text{LOE}}^{\text{NE}}$  and  $f^{\text{NE}}$  increases, as can be expected from any perturbation series expansion. The LOE gives physical results only when the electron-phonon coupling is such as  $2\pi\gamma_0^2/\Gamma_{L+R}\max[A_C(\omega)] < 0.5$ . Otherwise one gets non-physical results for the NE distribution, i.e.,  $f_{\text{LOE}}^{\text{NE}} > 1$  or  $f_{\text{LOE}}^{\text{NE}} < 0$ , as shown in Fig. 2(c). Such a behavior never occurs for the distribution  $f_{(1)}^{\text{NE,SC}}$  since it contains the proper renormalization. Therefore, in general, it is better to use an approximated distribution such as  $f_{(1)}^{\text{NE}}$  than the LOE.

Another important point to mention is shown in Figs. 2(b) and 2(c): The inelastic processes (see the sideband peak located around  $\omega \sim 0.3$ ) are only reproduced in the LOE when some form of self-consistency has been used, i.e., either in the form of  $f_{\text{LOE}}^{\text{NE}}[A_C^{\text{SC}}]$  or  $f_{\text{LOE}}^{\text{NE,SC}}$ . The LOE distribution calculated with the noninteracting spectral function  $A_C^0(\omega)$  is not able to reproduce such effects.

Finally, it should also be noted that all the self-consistent calculations including approximated distributions, such as  $f_{\text{LOE}}^{\text{NE}}$  or  $f_{(1)}^{\text{NE}}$ , converge much more quickly than the full calculation for  $f^{\text{NE}}$  (see Ref. [42] for more details). Such a numerical improvement is important for the calculations of more realistic and larger systems.

#### 4. Current and IETS signal

Figure 3 shows a typical result for the dynamical conductance  $G(V) = dI/dV$  obtained in the off-resonance transport regime. The current is calculated as in Ref. [4] using different approximations for the NE distribution function.

The conductance  $G(V)$  calculated with the approximated distribution  $f_{(1)}^{\text{NE}}$  provides a good representation of the conductance calculated with the exact distribution  $f^{\text{NE}}$ . The peak positions are well reproduced, but the amplitude of the conductance peaks is slightly larger with  $f_{(1)}^{\text{NE}}$ . This is due to the lack of full renormalization of  $f_{(1)}^{\text{NE}}$  in comparison to  $f^{\text{NE}}$ . The approximated distribution  $f_{(1)}^{\text{NE}}$  always gives a slightly larger electron population as shown in Fig. 2. We do not show the results obtained with  $f_{\text{LOE}}^{\text{NE}}$  since for coupling strengths  $\gamma_0/\omega_0 > 0.3$ ,  $f_{\text{LOE}}^{\text{NE}}$  gives nonphysical results as shown in Fig. 2(c).

The inelastic effects are best represented by the inelastic electron tunneling spectra (IETS) provided from the second

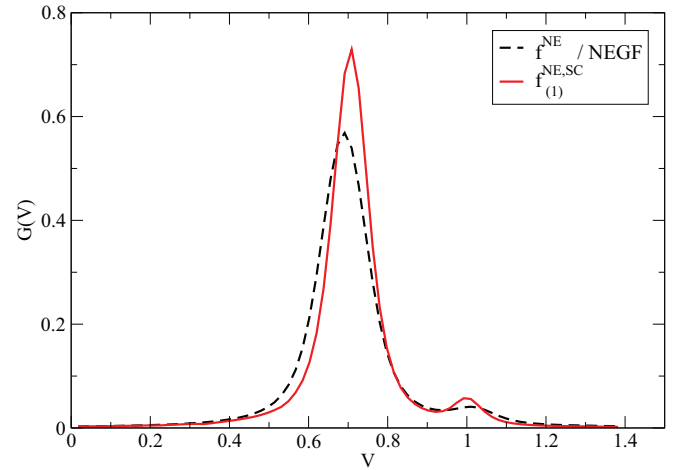


FIG. 3. (Color online) Dynamical conductance  $G(V) = dI/dV$  (in units of quantum of conductance  $G_0$ ) for the off-resonance regime ( $\varepsilon_0 = 0.70$ ).  $G(V)$  calculated with the approximated distribution  $f_{(1)}^{\text{NE}}$  (solid line) gives a good representation of the conductance calculated with the exact distribution  $f^{\text{NE}}$  (dashed line). The latter is strictly equivalent to NEGF calculations. The other parameters are  $\omega_0 = 0.3$ ,  $\gamma_0 = 0.10$ ,  $t_{0\alpha} = 0.19$ ,  $T_\alpha = 0.017$ ,  $\eta_L = 1$ ,  $\varepsilon_\alpha = 0$ ,  $\beta_\alpha = 2$ .

derivative of the current versus the applied bias. Figure 4 shows such a signal normalized by the conductance. As expected for the off-resonance regime [40,43–45], we obtain a peak in the IETS for the voltage threshold  $V \sim \omega_0$ . The exact IETS signal calculated with the distribution  $f^{\text{NE}}$  is well presented by the IETS calculated with the approximated distribution  $f_{(1)}^{\text{NE}}$ .

More interestingly, the results obtained with the LOE approximated distribution  $f_{\text{LOE}}^{\text{NE}}$  also give a good representation of the IETS signal, even for a coupling strength  $\gamma_0/\omega_0 = 0.4$ . We interpret such a behavior in the following manner: For

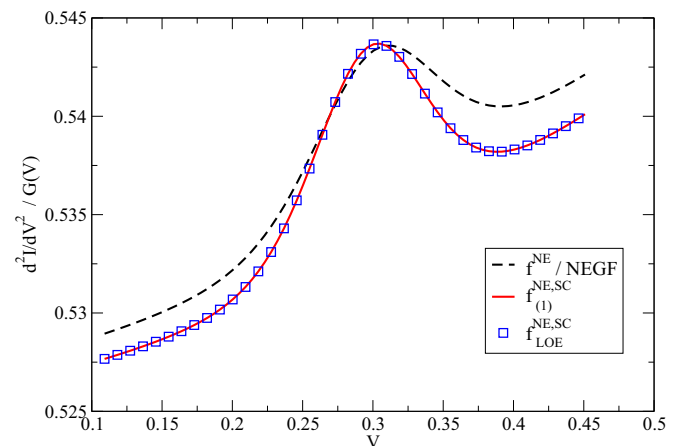


FIG. 4. (Color online) IETS signal  $d^2I/dV^2$ , normalized by the conductance  $G(V)$ , for the far-off-resonance regime ( $\varepsilon_0 = 3.70$ ). The IETS calculated with the approximated distributions  $f_{(1)}^{\text{NE}}$  (solid line) or even  $f_{\text{LOE}}^{\text{NE}}$  (squares) gives a good representation of the IETS calculated with the exact distribution  $f^{\text{NE}}$  (dashed line). The results obtained with  $f^{\text{NE}}$  are strictly equivalent to NEGF calculations. The other parameters are  $\omega_0 = 0.3$ ,  $\gamma_0 = 0.12$ ,  $t_{0L} = 0.45$ ,  $t_{0R} = 0.10$ ,  $T_\alpha = 0.017$ ,  $\eta_L = 1$ ,  $\varepsilon_\alpha = 0$ ,  $\beta_\alpha = 2$ .

small applied bias, where the transport is mostly tunneling and away from any resonant transport mechanisms, the LOE distribution  $f_{\text{LOE}}^{\text{NE}}$  is still realistic (i.e.,  $0 < f_{\text{LOE}}^{\text{NE}} < 1$ ) and quite close to the distribution  $f_{(1)}^{\text{NE}}$ . Hence both distributions provide similar results for the IETS signal. However, whenever the bias is large enough to include any resonances (main resonance or any phonon-sideband peak), the LOE distribution will provide a nonphysical behavior as shown in Fig. 2(c).

#### IV. TOWARDS MORE COMPLEX SYSTEMS

In order to extend the previous results to more realistic systems, we need to include several electron states and eventually several vibration modes in the central region. For that, we follow the same reasoning as in Sec. III, and consider the GFs as being matrices  $G_{nm}(\omega)$  in the electron level (or site) representation. The self-energies are also matrices in such a representation. We then define a different matrix for the NE distribution  $f_{nm}^{\text{NE}}$  as follows:

$$G_{C, nm}^<(\omega) = - \sum_l f_{nl}^{\text{NE}}(\omega) [G_{C, lm}^r(\omega) - G_{C, lm}^a(\omega)]. \quad (15)$$

With a few lines of algebra, we find that

$$\begin{aligned} \underline{f}^{\text{NE}}(\omega) &= \underline{G}_C^< [\underline{G}_C^a - \underline{G}_C^r]^{-1} \\ &= \underline{G}_C^r \underline{\Sigma}^< \underline{G}_C^a [\underline{G}_C^r [(\underline{G}_C^r)^{-1} - (\underline{G}_C^a)^{-1}] \underline{G}_C^a]^{-1} \\ &= \underline{G}_C^r \underline{\Sigma}^< [\underline{\Sigma}^a - \underline{\Sigma}^r]^{-1} (\underline{G}_C^r)^{-1} \\ &= \underline{G}_C^r (\underline{f}_0^{\text{NE}} \underline{\Gamma}_{L+R} - i \underline{\Sigma}_{\text{int}}^<) [\underline{G}_C^r (\underline{\Gamma}_{L+R} + \underline{\Gamma}_{\text{int}})]^{-1}, \end{aligned} \quad (16)$$

where  $\underline{\Gamma}_{\text{int}} = i(\underline{\Sigma}_{\text{int}}^> - \underline{\Sigma}_{\text{int}}^<)$ .

Equation (16) for  $f_{nm}^{\text{NE}}$  is more complicated than Eq. (4) because of the presence of the retarded GF terms which do not cancel in the general matrix form. Furthermore, the physical interpretation of  $\underline{f}^{\text{NE}}$  is more complicated. However, the diagonal matrix elements  $f_{nn}^{\text{NE}}$  represent the occupations of the level  $n$ , and the off-diagonal matrix elements represent some form of probability rate of transition between states.

It should be noticed that, however, all the functional analyses we have performed in Sec. III A and Appendix C still hold for the matrix case, i.e., the interaction self-energy is a functional of the spectral function and of the NE distribution. Furthermore,  $\underline{G}_C^r$  is also a functional of the spectral function,  $A_{C, nm}$ , which is now given in a matrix form, and  $G_{C, nm}^r(\omega) = \mathcal{H}[\pi A_{C, nm}(\omega)] - i\pi A_{C, nm}(\omega)$ .

Hence we can still use the functional property of the NE distribution, that is,  $\underline{f}^{\text{NE}} = \underline{f}^{\text{NE}}[\underline{f}^{\text{NE}}(\omega), \underline{A}_C(\omega)]$ , to devise a self-consistently iterative scheme to solve the problem. However, now, we have to take into account all the different matrix elements of the NE distribution and spectral functions.

We can choose for convenience that the coupling of the central region  $C$  to the lead  $\alpha$  is diagonal in the  $n, m$  representation:  $\Gamma_{\alpha nm} = \Gamma_{\alpha, n} \delta_{nm}$ . Hence the noninteracting NE distribution  $\underline{f}_0^{\text{NE}}$  is diagonal as well, with matrix elements:

$$f_{0, n}^{\text{NE}} = \frac{f_{L, n} \Gamma_{L, n}(\omega) + f_{R, n} \Gamma_{R, n}(\omega)}{\Gamma_{L, n}(\omega) + \Gamma_{R, n}(\omega)}, \quad (17)$$

with  $f_{\alpha, n} = f_{\alpha}(\omega = \epsilon_n)$  the population of the eigenvalue  $\epsilon_n$  of state  $n$  given from the statistics of the lead  $\alpha$ .

Furthermore, if the interaction is such that  $\underline{\Sigma}_{\text{int}}^<$  is also diagonal, the terms in  $G^r$  cancel in Eq. (16), and we end up with a set of  $n = 1, \dots, N$  equations such as Eq. (4) for  $f_{nn}^{\text{NE}}(\omega)$ , which need to be solved self-consistently for the  $n$  distributions and the  $n$  spectral functions  $A_{C, nn}(\omega)$ .

However, in the most general cases,  $\underline{\Sigma}_{\text{int}}^<$  is not diagonal, and one would need to solve the problem in a matrix form. For example, a generalization of the self-energies for electron-phonon coupling, given in Appendix B, would be [44–50]

$$\Sigma_{\text{int}, nm}^<(\omega) = \sum_{\nu} i \int \frac{du}{2\pi} D_{0, \nu}^<(u) \gamma_{\nu, np} G_{C, pq}^<(\omega - u) \gamma_{\nu, qm}, \quad (18)$$

where the coupling matrix elements  $\gamma_{\nu, np}$  correspond to an excitation of the vibration mode  $\nu$  (emission or absorption of a quantum) with electronic transition between state  $n$  and  $p$ .

We provide in Appendix E a specific example of a two-level model coupled to different vibration modes and show how to calculate the different matrix elements of  $\underline{f}^{\text{NE}}$ .

As far as we know, calculations for realistic systems (i.e., several electron levels and vibration modes) still need to be performed for the full range of NE and MB effects. NE distribution functions have been used in large systems but only for noninteracting cases or for cases where the interactions are treated in a mean-field manner [51]. The effects of NE and MB effects for  $e$ -ph coupled realistic systems have been considered, however, only at the level of a lowest-order expansion for the coupling, and in conjunction with some form of self-consistency [45, 48–50].

The really important point in the use of NE distributions for complex systems is that both the NE and MB effects are taken into account in the statistics of the finite size open quantum system (the central region  $C$ ). The NE distributions give the (fractional) electron population in the corresponding electronic levels in the presence of the NE conditions and for a given model of the MB effects (self-energies). One could envisage incorporating such NE statistics in density-functional-based codes able to deal with fractional occupation numbers for the corresponding Kohn-Sham states.

#### V. CONCLUSION

We have developed an alternative scheme to calculate the nonequilibrium (steady state) properties of open quantum systems. The method is based on the use of NE distribution and spectral functions. The method presents several advantages, but is strictly equivalent to conventional steady state NEGF calculations, when using the same level of approximations for the MB interaction. This is because there is a one-to-one correspondence with the NE distribution and spectral functions and the different GFs used in NEGF. The advantages of our method resides in the fact that the NE distribution and spectral functions have well behaved features for numerical applications, and that, for the single-level model, one works with purely real-number quantities.

Furthermore, our approach offers the possibility to introduce further approximations, not only at the level of the MB interaction (as in NEGF), but also at the level of the functional

forms used for the NE distributions. Introducing approximations at this level is important to reduce the computational cost of the method. For the model of the electron-phonon coupled system we have studied, such approximations provide a good representation of the full exact results, for either the NE distributions themselves or for physical measurable quantities such as the conductance and the IETS signal. An extension to systems consisting of several electron levels and several vibration modes has also been provided.

The concept of NE distribution functions also gives more direct physical information about the open quantum system, for example, in terms of depletion or accumulation of the electron population induced by the NE and MB effects. The NE distribution is also a useful concept to study other properties of the open quantum system, such as the NE charge susceptibility [5] and the NE fluctuation-dissipation relations [52].

We expect that such a method will be useful for the study of large and more realistic systems [53], such as single-molecule thermoelectric devices, as some approximated version of the NE distributions could be implemented in density-functional-based calculations [54].

#### ACKNOWLEDGMENTS

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#### APPENDIX A: INTERACTING VERSUS NONINTERACTING NE DISTRIBUTIONS

From the general expression of  $f^{\text{NE}}(\omega)$  in Eq. (4), there is no *a priori* reason for  $f^{\text{NE}}$  to be equal to the NE noninteracting distribution  $f_0^{\text{NE}}$ . In the very special cases where the interaction self-energy  $\Sigma_{\text{int}}^<$  follows the noninteracting statistics, i.e., in the sense that

$$\Sigma_{\text{int}}^< \stackrel{?}{=} -f_0^{\text{NE}}(\Sigma_{\text{int}}^r - \Sigma_{\text{int}}^a) = -f_0^{\text{NE}}(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<), \quad (\text{A1})$$

we obtain straightforwardly from Eq. (4) that  $f^{\text{NE}} = f_0^{\text{NE}}$ . Then all quantities, GFs and self-energies, follow the statistics given by the noninteracting case, as suggested in Ref. [57]. However, this is generally not true.

Indeed, even when the interactions are present only in the central region, it is not possible to ignore their indirect MB effects which spread throughout the systems. Such effects need to be incorporated into the local statistics. The latter cannot simply arise from the (noninteracting) lead statistics only.

For example, in the Anderson impurity model, the Kondo cloud generated by electron-electron interactions expands over more than the single site where the interaction is present. For electron-phonon interactions, when one performs a Lang-Firsov unitary transformation to diagonalize the interacting part of the Hamiltonian, one needs to keep the effects of such a transformation onto the effective coupling matrix elements between the (now diagonal) central region and the leads' Hamiltonians. In simple words, one could say that the electron-phonon interaction is now crossing at the contacts between the central region and the leads. Therefore, there is

no reason to assume that the corresponding statistics would be given by the noninteracting one.

Moreover, there are clear indications from numerical calculations that  $f^{\text{NE}} \neq f_0^{\text{NE}}$ . This has been shown for electron-electron interactions (for example, see Fig. 3 in Ref. [37]) and for electron-phonon interactions (for example, see Figs. 5 and 6 in Ref. [58], Fig. 6 in Ref. [59], and Fig. 7 in Ref. [60]). We also provide a few examples in Sec. III C 2.

We can also convince ourselves that generally  $f^{\text{NE}} \neq f_0^{\text{NE}}$  by considering the following example for electron-phonon interactions. The lowest-order diagram for which the self-energies  $\Sigma_{\text{int}}^{\lessgtr}$  are not vanishing is the Fock diagram [39,40] (see Appendix B):

$$\Sigma_{\text{int}}^{F,\lessgtr}(\omega) = \gamma_0^2 [N_{\text{ph}} G_C^{\lessgtr}(\omega \mp \omega_0) + (N_{\text{ph}} + 1) G_C^{\lessgtr}(\omega \pm \omega_0)]. \quad (\text{A2})$$

One can use the ratio  $\Sigma_{\text{int}}^{F,>} / \Sigma_{\text{int}}^{F,<}$  to define a distribution function

$$f_{\text{int}}^{\text{NE}}(\omega) = \left( 1 - \frac{\Sigma_{\text{int}}^{F,>}(\omega)}{\Sigma_{\text{int}}^{F,<}(\omega)} \right)^{-1}, \quad (\text{A3})$$

such as  $\Sigma_{\text{int}}^< = -f_{\text{int}}^{\text{NE}}(\Sigma_{\text{int}}^> - \Sigma_{\text{int}}^<) = -f_{\text{int}}^{\text{NE}}(\Sigma_{\text{int}}^r - \Sigma_{\text{int}}^a)$ . At low temperature,  $N_{\text{ph}} = 0$  and the ratio

$$\frac{\Sigma_{\text{int}}^{F,>}}{\Sigma_{\text{int}}^{F,<}} = \frac{G_C^>(\omega - \omega_0)}{G_C^<(\omega + \omega_0)} \quad (\text{A4})$$

defines a distribution  $f_{\text{int}}^{\text{NE}}$  which is clearly different from  $f_0^{\text{NE}}$ . Indeed, if  $f_{\text{int}}^{\text{NE}} = f_0^{\text{NE}}$ , one has  $\Sigma_{\text{int}}^{F,>} / \Sigma_{\text{int}}^{F,<} = (f_0^{\text{NE}} - 1) / f_0^{\text{NE}}$ , which is not possible from the definition of Eq. (A4).

To further convince ourselves, let us assume that  $G_C^{\lessgtr}$  were following the distribution  $f_0^{\text{NE}}$ . Then from Eq. (A4), we would have

$$\begin{aligned} \frac{\Sigma_{\text{int}}^{F,>}}{\Sigma_{\text{int}}^{F,<}} &= \frac{G_C^>(\omega - \omega_0)}{G_C^<(\omega + \omega_0)} = \frac{f_0^{\text{NE}}(\omega - \omega_0) - 1}{f_0^{\text{NE}}(\omega + \omega_0)} \frac{A_C(\omega - \omega_0)}{A_C(\omega + \omega_0)} \\ &\neq \frac{f_0^{\text{NE}}(\omega) - 1}{f_0^{\text{NE}}(\omega)}, \end{aligned} \quad (\text{A5})$$

where  $A_C(\omega)$  is the spectral function of the central region  $C$ . The inequality in Eq. (A5) holds even for the symmetric electron-hole case [61].

Hence, we can safely conclude that, in the most general cases, the two distribution functions  $f^{\text{NE}}$  and  $f_0^{\text{NE}}$  differ from each other.

#### APPENDIX B: THE ELECTRON-PHONON SELF-ENERGIES

The electron-phonon self-energies in the central region  $C$  are calculated within the self-consistent Born approximation. The details of the calculations are reported elsewhere [39,40], so we briefly recall the different expressions for the self-energies  $\Sigma_{\text{int}}^x(\omega) = \Sigma_C^{H,x}(\omega) + \Sigma_C^{F,x}(\omega)$  with

$$\Sigma_C^{H,r} = \Sigma_C^{H,a} = 2 \frac{\gamma_0^2}{\omega_0} \int \frac{d\omega'}{2\pi} i G_C^<(\omega') = -2 \frac{\gamma_0^2}{\omega_0} \langle n_C \rangle, \quad (\text{B1})$$



with  $\langle n_C \rangle = -i \int d\omega/2\pi G_C^<(\omega)$  and

$$\Sigma_C^{F,\lessgtr}(\omega) = i\gamma_0^2 \int \frac{du}{2\pi} D_0^{\lessgtr}(u) G_C^{\lessgtr}(\omega - u), \quad (\text{B2})$$

and

$$\begin{aligned} \Sigma_C^{F,r}(\omega) &= i\gamma_0^2 \int \frac{du}{2\pi} D_0^r(\omega - u)(G_C^<(u) + G_C^r(u)) \\ &\quad + D_0^<(\omega - u)G_C^r(u), \end{aligned} \quad (\text{B3})$$

with the usual definitions for the bare vibron GF  $D_0^{\lessgtr}$ :

$$\begin{aligned} D_0^{\lessgtr}(\omega) &= -2\pi i [N_{\text{ph}}\delta(\omega \mp \omega_0) + (N_{\text{ph}} + 1)\delta(\omega \pm \omega_0)], \\ D_0^r(\omega) &= \frac{1}{\omega - \omega_0 + i0^+} - \frac{1}{\omega + \omega_0 + i0^+}, \end{aligned} \quad (\text{B4})$$

where  $N_{\text{ph}}$  is the averaged number of excitations in the vibration mode of frequency  $\omega_0$  given by the Bose-Einstein distribution at temperature  $T_{\text{ph}}$ .

We are mostly interested to see how the inelastic effects are reproduced by our method based on the NE distribution. Therefore we ignore the contribution of the static part of the interaction, i.e., the Hartree-like self-energy  $\Sigma_C^{H,r/a}$ , in the calculations. Note, however, that since the NE distribution is defined from the lesser and greater components of the interaction self-energies, the Hartree-like component is not relevant for the calculation of  $f^{\text{NE}}$ .

### APPENDIX C: FUNCTIONAL FORMS OF THE NE DISTRIBUTION

We analyze in this Appendix the dependence of  $f^{\text{NE}}$  on the MB effects using a conventional diagrammatic NE approach for the interactions. The lowest-order nonvanishing lesser and greater self-energies have the form of a convolution product of the following type:

$$\Sigma_{\text{int}}^{\lessgtr}(\omega) = i \int \frac{du}{2\pi} \mathcal{B}^{\lessgtr}(u) G_C^{\lessgtr}(\omega - u), \quad (\text{C1})$$

where  $\mathcal{B}(\omega)$  is related to a boson propagator.

For electron-phonon interactions,  $\mathcal{B}(\omega)$  is given by  $\mathcal{B}(\omega) = \gamma_0^2 D(\omega)$ , where  $D(\omega)$  is the phonon propagator and  $\gamma_0$  is the electron-phonon coupling constant. Different levels of approximation can be used by considering the bare phonon propagator  $D_0(\omega)$ , or a partially dressed phonon propagator  $\mathcal{D}_0(\omega)$ , or the fully dressed phonon propagator  $\mathcal{D}(\omega)$ .

For electron-electron interactions,  $\mathcal{B}(\omega)$  is the screened Coulomb interaction  $W(\omega)$  in which the screening is obtained according to different levels of approximation. We describe a few of them in the following.

*Electron-phonon interaction.* When dealing with the bare phonon, the lesser interaction self-energy becomes

$$\begin{aligned} \Sigma_{\text{int}}^<(\omega) &= i\gamma_0^2 \int \frac{du}{2\pi} D_0^<(\omega - u) G_C^<(u) \\ &= -\gamma_0^2 \int du D_0^<(\omega - u) f^{\text{NE}}(u) A_C(u). \end{aligned} \quad (\text{C2})$$

Clearly such a self-energy is a functional of the NE distribution  $f^{\text{NE}}(u)$  and of the spectral function  $A(u)$ . One obtains a similar result for the greater self-energy  $\Sigma_{\text{int}}^>(\omega)$ .

For the partially dressed  $\mathcal{D}_0$  or the fully dressed phonon propagator  $\mathcal{D}$ , we have the following expressions for the propagator:  $\mathcal{D}_0(\omega) = D_0(\omega) + D_0(\omega)\gamma_0^2 P(\omega)D_0(\omega)$  or  $\mathcal{D}(\omega) = [D_0(\omega)^{-1} - \gamma_0^2 P(\omega)]$  with  $P(\omega)$  being the polarization function. At the lowest order, the polarization is given by the electron-hole bubble diagram and its lesser and greater components are

$$\begin{aligned} P^{\lessgtr}(\omega) &= -i \int \frac{du}{2\pi} G_C^{\lessgtr}(u) G_C^{\gtrless}(u - \omega) \\ &= -i2\pi \int du f^{\text{NE}}(u)[1 - f^{\text{NE}}(u - \omega)] \\ &\quad \times A_C(u)A_C(u - \omega), \end{aligned} \quad (\text{C3})$$

which is again a functional of  $f^{\text{NE}}$  and  $A_C$ . Therefore we find that for any phonon propagator, we have  $\Sigma_{\text{int}}^< = \Sigma_{\text{int}}^<[f^{\text{NE}}, A_C]$ .

*Electron-electron interaction.* The screened Coulomb interaction  $W(\omega) = v_q/\epsilon^{-1}(\omega, q)$  can be calculated within a different level of approximation for the dielectric function  $\epsilon^{-1}(\omega, q)$  ( $v_q$  is the Fourier  $q$  component of the bare Coulomb interaction).

In the plasmon-pole approximation [62,63], we have  $\epsilon^{-1}(\omega, q) = 1 + \omega_p^2/(\omega^2 - \omega_q^2)$ , where  $\omega_p$  is the bulk plasmon energy and  $\omega_q$  the plasmon dispersion relation. The dynamic part of the screened Coulomb potential  $W(\omega) - v$  can be rewritten as

$$v_q(\epsilon^{-1}(\omega, q) - 1) = \frac{v_q\omega_p^2}{2\omega_q} \frac{2\omega_q}{\omega^2 - \omega_q^2} = \gamma_p^2 B_p(\omega, q), \quad (\text{C4})$$

which involves a coupling constant  $\gamma_p$  and the bosonic propagator  $B_p(\omega)$  of the plasmon modes. This corresponds to the similar case of the bare phonon propagator described above. Using the same reasoning, we find that the interacting self-energy  $\Sigma_{\text{int}}^<$  is a functional of  $f^{\text{NE}}$  and  $A_C$ .

Within the *GW* approximation [62–67], the screened Coulomb interaction is given by  $W(\omega) = v + vP(\omega)W(\omega)$ . This expression is a formally equivalent to the case of the fully dressed phonon propagator since  $\mathcal{D}(\omega) = [D_0(\omega)^{-1} - \gamma_0^2 P(\omega)] = D_0(\omega) + D_0(\omega)\gamma_0^2 P(\omega)\mathcal{D}(\omega)$ . Hence, applying the previous analysis, we find again that  $\Sigma_{\text{int}}^< = \Sigma_{\text{int}}^<[f^{\text{NE}}, A_C]$ .

*Vertex corrections and higher-order diagrams.* We can also consider higher-order diagrams for the electron-phonon and electron-electron, as well as vertex corrections to build more elaborate self-energies. From our earlier work [39,40,63], it can be seen from the expressions of the second-order and vertex correction diagrams that the self-energies  $\Sigma_{\text{int}}^{\lessgtr}$  can always be expressed as a functional of the NE distribution  $f^{\text{NE}}(\omega)$  and of the spectral function  $A_C(\omega)$  [68].

### APPENDIX D: LOWEST-ORDER EXPANSION FOR THE CURRENT

For the two-terminal quantum devices we consider, the current  $I(V)$  is given by the famous Meir and Wingreen expression [69]:

$$\begin{aligned} I &= \frac{ie}{h} \int d\omega \text{Tr} \{ [f_L(\omega)\Gamma_L - f_R(\omega)\Gamma_R] [G_C^r(\omega) - G_C^a(\omega)] \\ &\quad + [\Gamma_L(\omega) - \Gamma_R(\omega)] G_C^<(\omega) \}, \end{aligned} \quad (\text{D1})$$

where we recall that  $\Gamma_\alpha(\omega)$  is the spectral function of the lead  $\alpha$  self-energy, i.e.,  $\Gamma_\alpha = i(\Sigma_\alpha^r - \Sigma_\alpha^a) = i(\Sigma_\alpha^> - \Sigma_\alpha^<)$ .

For the single-impurity model, the trace drops off and one deals with functions only. Using the definitions  $2\pi A_C = i(G_C^r - G_C^a)$  and  $G_C^< = -f^{\text{NE}}(G_C^r - G_C^a)$ , we obtain

$$I = \frac{2\pi e}{h} \int d\omega (f_L \Gamma_L - f_R \Gamma_R) A_C - (\Gamma_L - \Gamma_R) f^{\text{NE}} A_C. \quad (\text{D2})$$

The lowest-order expansion of the current, in terms of elastic and inelastic processes, is obtained by introducing the approximated form Eq. (12) for the NE distribution. The current is built on two contributions  $I = I_{\text{el}} + I_{\text{inel}}$  with

$$\begin{aligned} I_{\text{el}} &= \frac{2\pi e}{h} \int d\omega (f_L \Gamma_L - f_R \Gamma_R) A_C - (\Gamma_L - \Gamma_R) f_0^{\text{NE}} A_C \\ &= \frac{e}{h} \int d\omega (f_L - f_R) \frac{2\Gamma_L \Gamma_R}{\Gamma_{L+R}} 2\pi A_C, \end{aligned} \quad (\text{D3})$$

where the second line is simply obtained from the definition of the noninteraction NE distribution Eq. (2). We can identify  $I_{\text{el}}$  in Eq. (D3) as a Landauer-like current expression with the transmission given by the usual formula  $T(\omega) = \text{Tr}[\Gamma_L(\omega) G_C^r(\omega) \Gamma_R(\omega) G^a(\omega)] + [\Gamma_R(\omega) G_C^r(\omega) \Gamma_L(\omega) G^a(\omega)] \equiv 2\Gamma_L \Gamma_R / \Gamma_{L+R} 2\pi A_C(\omega)$ . This is a purely elastic transmission when the GFs or  $A_C(\omega)$  are calculated in the absence of an interaction. In the presence of an interaction, we are dealing with elastic transport with renormalized GFs [4].

The second contribution to the current is given by

$$\begin{aligned} I_{\text{inel}} &= -\frac{e}{h} (2\pi \gamma_0)^2 \int d\omega \frac{\Gamma_L - \Gamma_R}{\Gamma_{L+R}} \\ &\quad \times \{ A_C(\omega + \omega_0) f_0^{\text{NE}}(\omega + \omega_0) A_C(\omega) [1 - f_0^{\text{NE}}(\omega)] \\ &\quad - A_C(\omega) f_0^{\text{NE}}(\omega) A_C(\omega - \omega_0) [1 - f_0^{\text{NE}}(\omega - \omega_0)] \}. \end{aligned} \quad (\text{D4})$$

This is simply the lowest-order inelastic contribution to the current, corresponding to vibron emission by the electron and hole. When Eq. (D4) is recast in terms of the Fermi distributions  $f_L$  and  $f_R$  entering the definition of  $f_0^{\text{NE}}$ , one recovers the lowest-order expansion results obtained from scattering theory [70–72] and from NEGF [45,46,50,73] if the spectral function  $A_C$  is calculated in the absence of an interaction.

The important point here is that our results are obtained in a rather straightforward manner by using the concept of NE distribution. They are equivalent to others when working within with same degree of approximation for the interaction self-energy. However, with the use of approximated forms for the NE distribution, we can still perform self-consistent calculations, which go beyond second-order perturbation theory.

## APPENDIX E: A TWO-LEVEL MODEL

In this Appendix, we provide an example for a model of the central region consisting of two levels  $i, j = 1, 2$  with two different kinds of  $e$ -ph coupling, a local Holstein-like coupling

on each site and an off-diagonal Su-Schrieffer-Heeger-like coupling between the two levels.

The interaction self-energies are nondiagonal  $2 \times 2$  matrices with elements  $\Sigma_{ij}^{\lessgtr}(\omega)$  given (in the limit of low temperature) by

$$\begin{aligned} \Sigma_{\text{int},11}^{\lessgtr}(\omega) &= \gamma_{0,1}^2 G_{C,11}^{\lessgtr}(\omega \mp \omega_1) \\ \Sigma_{\text{int},12}^{\lessgtr}(\omega) &= \gamma_{0,\text{od}}^2 G_{C,12}^{\lessgtr}(\omega \mp \omega_{\text{od}}) \\ \Sigma_{\text{int},21}^{\lessgtr}(\omega) &= \gamma_{0,\text{od}}^2 G_{C,21}^{\lessgtr}(\omega \mp \omega_{\text{od}}) \\ \Sigma_{\text{int},22}^{\lessgtr}(\omega) &= \gamma_{0,2}^2 G_{C,22}^{\lessgtr}(\omega \mp \omega_2), \end{aligned} \quad (\text{E1})$$

where  $\omega_i$  and  $\gamma_{0,i}$  are the energy and coupling constants for the local  $e$ -ph interaction on level  $i = 1, 2$  and  $\omega_{\text{od}}$  and  $\gamma_{0,\text{od}}$  are the corresponding quantities for the nonocal  $e$ -ph interaction between level 1 and 2.

For simplicity we consider the coupling to the lead is diagonal, i.e.,  $\Gamma_{L+R,ij} = \Gamma_i \delta_{ij}$  and therefore the noninteracting NE distribution matrix  $f_{-0}^{\text{NE}}$  is also diagonal, with elements  $f_{0,i}^{\text{NE}}$  given by Eq. (17).

We focus in the following on the LOE of  $f^{\text{NE}}$ . This approximation still shows how the different components of the NE distribution matrix are obtained in the presence of a nondiagonal interaction self-energy.

Following the derivation given in Sec. III C 3, Eq. (16) can be recast as

$$\begin{aligned} \underline{f}^{\text{NE}}(\omega) &\sim \underline{G}_C^r (f_0^{\text{NE}} \Gamma_{L+R} - i \underline{\Sigma}_{\text{int}}^<) \Gamma_{L+R}^{-1} (1 - \Gamma_{L+R}^{-1} i \underline{\Sigma}_{\text{int}}^>-<) \\ &\quad \times (\underline{G}_C^r)^{-1} \\ &\sim \underline{G}_{0,C}^r (f_0^{\text{NE}} - i \underline{\Sigma}_{\text{int}}^< \Gamma_{L+R}^{-1} - f_0^{\text{NE}} \Gamma_{L+R}^{-1} i \underline{\Sigma}_{\text{int}}^>-<) \\ &\quad \times (\underline{G}_{0,C}^r)^{-1}, \end{aligned} \quad (\text{E2})$$

where we kept only the lowest-order terms,  $\underline{\Sigma}_{\text{int}}^>-<$  is a contraction for  $\underline{\Sigma}_{\text{int}}^>-< = \underline{\Sigma}_{\text{int}}^> - \underline{\Sigma}_{\text{int}}^<$ , and  $\underline{G}_{0,C}^r$  is the noninteracting GF of the region  $C$ . Such a GF is diagonal with elements  $G_{0,i}^r(\omega)$  in the two-level representation. Hence we obtain the following LOE for  $\underline{f}^{\text{NE}}$ :

$$\underline{f}_{\text{LOE}}^{\text{NE}}(\omega) = \begin{bmatrix} F_{11}(\omega) & r(\omega) F_{11}(\omega) \\ F_{21}(\omega)/r(\omega) & F_{22}(\omega) \end{bmatrix}, \quad (\text{E3})$$

where  $r(\omega)$  is ratio  $r = G_{0,1}^r(\omega)/G_{0,2}^r(\omega)$  and  $F_{ij}$  are the matrix elements of

$$\underline{F} = \underline{f}_0^{\text{NE}} - i \underline{\Sigma}_{\text{int}}^< \Gamma_{L+R}^{-1} - \underline{f}_0^{\text{NE}} \Gamma_{L+R}^{-1} i \underline{\Sigma}_{\text{int}}^>-<. \quad (\text{E4})$$

By the definition Eq. (15), the matrix elements of  $\underline{G}_C^{\lessgtr}$  entering the definition of the self-energies  $\underline{\Sigma}_{\text{int}}^{\lessgtr}$  depend on both the diagonal and off-diagonal elements of  $\underline{f}^{\text{NE}}$ .

However, at the LOE, we substitute  $\underline{f}^{\text{NE}}$  with the noninteracting distribution  $\underline{f}_0^{\text{NE}}$  which is diagonal. Hence we have

$$\begin{aligned} G_{C,ij}^<(\omega) &= i2\pi f_{0,i}^{\text{NE}}(\omega) A_{C,ij}(\omega) \\ G_{C,ij}^>(\omega) &= i2\pi (f_{0,i}^{\text{NE}}(\omega) - 1) A_{C,ij}(\omega). \end{aligned} \quad (\text{E5})$$

After substitution into the definition of the self-energy Eq. (E1), we obtain from Eq. (E4) the following matrix

elements of  $\underline{F}$ :

$$F_{ii}(\omega) = f_{0,i}^{\text{NE}}(\omega) + \frac{2\pi\gamma_{0,i}^2}{\Gamma_i} \left\{ A_{C,ii}(\omega + \omega_i) f_{0,i}^{\text{NE}}(\omega + \omega_i) \right. \\ \times [1 - f_{0,i}^{\text{NE}}(\omega)] - A_{C,ii}(\omega - \omega_i) [1 - f_{0,i}^{\text{NE}}(\omega - \omega_i)] \\ \left. \times f_{0,i}^{\text{NE}}(\omega) \right\}, \quad (\text{E6})$$

for the diagonal elements ( $i = 1, 2$ ) and for the off-diagonal elements:

$$F_{12}(\omega) = \frac{2\pi\gamma_{\text{od}}^2}{\Gamma_1} \left( \left[ \frac{\Gamma_1}{\Gamma_2} - f_{0,1}^{\text{NE}}(\omega) \right] f_{0,1}^{\text{NE}}(\omega + \omega_{\text{od}}) \right. \\ \times A_{C,12}(\omega + \omega_{\text{od}}) - A_{C,12}(\omega - \omega_{\text{od}}) \\ \left. \times [1 - f_{0,1}^{\text{NE}}(\omega - \omega_{\text{od}})] f_{0,1}^{\text{NE}}(\omega) \right). \quad (\text{E7})$$

The matrix element  $F_{21}$  is obtained from the expression of  $F_{12}$  by swapping the indices  $1 \leftrightarrow 2$ .

From Eqs. (E6), (E7), and (E3), we can see that the diagonal elements  $f_{\text{LOE},ii}^{\text{NE}}$  are real and given by an expression similar to the result Eq. (12) obtained for the single-level model. The off-diagonal elements  $f_{\text{LOE},ij}^{\text{NE}}$  acquire an imaginary part via the presence of the ratio  $r(\omega)$ . In some cases, such an imaginary can be negligible or even vanishing.

The interesting point in the LOE is that each matrix element  $f_{\text{LOE},ij}^{\text{NE}}$  is to be determined self-consistently with the corresponding matrix element  $A_{C,ij}$  of the spectral function. There is no mixing between the different  $A_{C,ij}$  and  $f_{\text{LOE},ij}^{\text{NE}}$ .

Obviously, beyond the LOE, there will be some mixing between the different matrix elements of the NE distribution and the spectral function, since  $\underline{G}_C^r$  is generally not diagonal and  $\underline{G}_C^<$  is given by

$$\underline{G}_C^< = i2\pi \begin{bmatrix} f_{11}^{\text{NE}} & f_{12}^{\text{NE}} \\ f_{21}^{\text{NE}} & f_{22}^{\text{NE}} \end{bmatrix} \begin{bmatrix} A_{C,11} & A_{C,12} \\ A_{C,21} & A_{C,22} \end{bmatrix}. \quad (\text{E8})$$

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