Time-dependent theory of nonlinear response and current fluctuations

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A general nonlinear response theory is derived for an arbitrary time-dependent Hamiltonian, not necessarily obeying time-reversal symmetry. We consider the application of this theory to a multiterminal mesoscopic system with arbitrary interactions and time-dependent voltages. This allows us to obtain a generalized Kubo-type formula. We derive a microscopic expression for the finite frequency differential conductance matrix, which preserves current conservation and gauge invariance. We exploit this result to show that the asymmetric part of the current fluctuation matrix at finite frequency obeys a generalized time-dependent fluctuation-dissipation theorem. In the stationary regime, this theorem provides a common explanation for the asymmetry of the excess noise with respect to positive and negative frequencies that has been obtained in several systems as a consequence of nonlinearity. It also explains the origin of the unexpected negative sign of the excess noise. Finally, we apply these general results to the case of a tunnel junction and obtain a nonperturbative out-of-equilibrium link between conductance and current fluctuations. We also derive a universal property of the finite frequency noise in the perturbative regime.

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I. INTRODUCTION

Linear response theory is a cornerstone of quantum theory. It allows one to derive extremely useful formulas such as the Kubo formula[1] or the fluctuation-dissipation theorem (FDT),[2–4] which are extensively used in all fields. It has been commonly believed that the validity of the Kubo formula is essentially limited to the linear regime. This has motivated the search for alternative methods to deal with correlated systems[5,6] or weak nonlinearities.[7] In this work we present an elegant and simple proof for a generalized Kubo formula that is valid for systems which have both a nonlinear behavior and a time-dependent Hamiltonian, from which we obtain a time-dependent, out-of-equilibrium FDT relation.

The derivation of these results is based on a formal and exact evaluation of the evolution of operators under the action of time-dependent external parameters, without making any assumption on the system. These results can thus be used indifferently in atomic physics, quantum chemistry, condensed matter physics, etc. Here we choose to illustrate the power of this approach by deriving new relations of general validity in multiterminal mesoscopic systems with arbitrary time dependence of the electrochemical potentials at the terminals. Multiply connected structures have been of great interest in quantum transport[8,9] because they can reveal more information compared to the two-terminal setup. This is, for instance, the case of single- or two-particle quantum interference in Aharonov-Bohm geometries,[10] Hanbury-Brown–Twiss setups[11,12] related to statistics and entanglement,[13] and Mach-Zehnder interferometers.[14] Quantum Hall edge states are particularly suited for performing quantum optics with electrons. It is, however, important to go beyond the independent electron picture, as Coulomb interactions have to be taken into account: they intervene either at integer filling or, in a more fundamental way, within the fractionnal quantum Hall effect (FQHE). In the latter case, for instance, three-terminal geometries have been proposed in order to probe fractional statistics,[15] which require one to consider out-of-equilibrium transport.

In this paper, we give a microscopic expression for the differential out-of-equilibrium conductance matrix with minimal assumptions on the system under study, and ensuring current conservation and gauge invariance. We thereby provide a convenient general framework to describe the time-dependent behavior of interacting multiterminal systems in a single, uniform formalism. This contrasts markedly with previous approaches in which *ad hoc* approximations often had to be made, depending on the system and questions under investigation. The formalism should prove to be useful both for the stationary regime and for time-dependent Hamiltonians. Our approach allows us to consider not only ac voltages, and thus photo-assisted transport,[16–20] but also arbitrary time dependence of the voltages. Therefore this approach should allow one to study the injection of electrons on demand,[21–23] classical sources of noise, pumping,[8,24] or mixing setups,[25] where the potentials in the terminals have different periods, etc.

We can also consider spontaneous generation under a dc bias, such as finite frequency (FF) noise,[11] or combine both, for instance, by applying time-dependent voltages and considering FF current fluctuations, which in this situation depend on two frequencies[26] and form a matrix containing both autocorrelations and cross correlations. By using our out-of-equilibrium and time-dependent Kubo formula, we show that the asymmetric part of this matrix obeys a general time-dependent out-of-equilibrium FDT. This FDT goes well beyond that obtained in the stationary regime for linear systems,[27] and for the nonlinear scalar conductivity[28,29] (obtained for antennas or one-dimensional systems), in particular, it is not restricted to stationary situations. We will nevertheless discuss its consequences in the stationary case, where we derive universal properties of the FF current fluctuations matrix.

An application showing the utility of the present formalism has been performed in quantum wires and carbon nanotubes described by a Tomonoga-Luttiger liquid (TLL),[30] where the FF conductance, depending on the dc voltage, has been analyzed in these prototypical strongly correlated systems. A recent investigation has been achieved in a quantum dot in Ref. 31. Here we apply our nonlinear response theory to a tunnel junction between arbitrary correlated systems in an arbitrary dimension subject to a time-dependent voltage and tunneling amplitude, not necessarily weak, which is contrary to most previous work on tunnel junctions.[32,33] We will derive a general Kubo-type formula for the differential FF conductance where we show the presence of a diamagnetic term, which is rather related to the tunneling Hamiltonian. In particular, this formula applies also to weak impurities in a one-dimensional system, as that studied in the nonlinear regime,[30] and extends that obtained in the linear regime in Ref. 22.

II. GENERALIZED NONLINEAR RESPONSE THEORY

For generality, we consider a system with a time-dependent Hamiltonian $\mathcal{H}(t)$, which includes arbitrary interactions or disorder, and does not necessarily obey time-reversal symmetry. $\mathcal{H}(t)$ depends on a set of time-dependent parameters generically denoted by $X(t')$. For any operator $\hat{O}(t)$, we denote its average $O(t) = \mathrm{Tr}[\rho \hat{O}(t)]$, taken in the presence of $\mathcal{H}(t)$ and with an initial density matrix ρ, which need not be thermal. The evolution of $O(t)$ under an arbitrary time evolution of the parameter $X(t')$ can be formally expressed as

$$O(t) = \int_0^1 d\varepsilon \int dt' \frac{\delta O_\varepsilon(t)}{\delta X(t')} X(t'),$$

where $O_\varepsilon(t)$ is computed replacing $X \to \varepsilon X$, and $\frac{\delta O_\varepsilon(t)}{\delta X(t')}$ is its functional derivative with respect to $X(t')$, playing the role of a generalized susceptibility for a nonlinear system.

To obtain $\frac{\delta O(t)}{\delta X(t')}$ (for $\varepsilon = 1$, but the derivation is obviously similar for other values of ε), we split the Hamiltonian into the part that does not depend on X, denoted by $\mathcal{H}_0(t)$, and the remaining part that depends on $X(t)$: $\mathcal{H}(t) = \mathcal{H}_0(t) + \mathcal{H}_1[t, X(t)]$.[34] We switch to the interaction picture where $\mathcal{H}_1$ is viewed as the interaction Hamiltonian. Then, $\hat{O}^{\mathrm{int}}(t) = U_0(-\infty, t)\hat{O}(t)U_0(t, -\infty)$, where $i\hbar\partial_t U_0(t, -\infty) = \mathcal{H}_0(t)U_0(t, -\infty)$. Even though this is not strictly necessary, we prefer to exploit the Keldysh formulation[35] to make our argument more compact. The Keldysh time contour has two branches labeled by $\eta = \pm$, going from $-\infty$ to ∞ on the upper one and inversely on the lower one. T_K is the Keldysh ordering operator, which makes time (antitime) ordering on the upper (lower) contour, while operators labeled with a minus sign (−) are placed on the left of these labeled with a plus sign (+): $\langle T_K A^+(t) B^-(t')\rangle = \langle \hat{B}(t')\hat{A}(t)\rangle$. $\hat{O}$ can be labeled indifferently by $\eta = +$ or $-$ to express its average. The functional derivative with respect to $X(t')$ reads

$$\begin{aligned}\frac{\delta O(t)}{\delta X(t')} &= \frac{\delta}{\delta X(t')}\left\langle T_K \hat{O}^+(t) e^{-\frac{i}{\hbar}\int_{-\infty}^{\infty}\sum_\eta \eta \mathcal{H}_1^\eta}\right\rangle \\ &= \frac{-i}{\hbar}\theta(t-t')\left\langle\left[\hat{O}(t), \frac{\delta \mathcal{H}}{\delta X(t')}\right]\right\rangle + \delta(t-t')\left\langle \frac{\partial \hat{O}}{\partial X}(t)\right\rangle,\end{aligned} \tag{1}$$

where we have used $\sum_\eta \eta \langle T_K A^+(t) B^\eta(t')\rangle = \theta(t-t')\langle[\hat{A}(t), \hat{B}(t')]\rangle$, and the fact that $\mathcal{H}_0$ does not depend on X. In the last term, we have further assumed that $\hat{O}$ does not depend on the time derivatives $\partial_t X, \partial_{t^2} X, \ldots$, in order to avoid a cumbersome expression.

III. APPLICATION TO TRANSPORT AND FLUCTUATIONS IN ARBITRARY TIME-DEPENDENT MULTITERMINAL MESOSCOPIC SYSTEMS

Now we apply this formula to a mesoscopic system described by a Hamiltonian $\mathcal{H}_0(t)$, which can, for instance, include time-dependent scatterers (see Fig. 1). It is connected to N reservoirs with electrochemical potential $\mu_n(t) = eV_n(t)$ and a total charge operator Q_n; thus the outgoing current reads

$$I_n = \partial_t Q_n.$$

The system is also coupled to a gate whose role could be played, for instance, by the ground. Extension to many gates is straightforward. One can view the system and the gate as a capacitor, and the current measured on the gate side is given by $I_0 = \partial_t Q_0$. On the other hand, by charge conservation, the total charge of the system and reservoirs is zero, as they do not exchange electrons with the gate:

$$\sum_{n=1}^{N} Q_n + Q_0 = 0, \tag{2}$$

and thus Kirchoff's law is ensured as

$$\sum_{n=0}^{N} I_n = 0. \tag{3}$$

These last two equations assume that the plasma frequency is high enough, otherwise they would need to be modified to take into account local charge density in the leads. We discuss now the coupling to reservoirs. These are defined as in the standard scattering approach, where inelastic processes are efficient enough to ensure a quasiequilibrium state. This requires the inelastic time to be smaller than the characteristic time scale of variations of $\mu_n(t)$. Their modeling can be chosen to make the microscopic calculation convenient. For instance, one could introduce intermediate ideal leads between the system and the reservoirs, in which case a global Hamiltonian $\mathcal{H}_0$ would include a noninteracting Hamiltonian for the leads,

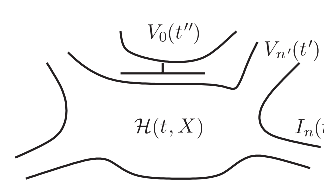

FIG. 1. A mesoscopic system coupled to many terminals, including a gate, with arbitrary time dependence of their electrochemical potentials. The time-dependent Hamiltonian $\mathcal{H}$ includes arbitrary interactions or disorder and can depend on other parameters $X(t')$ in a nonlocal and nonlinear way. The differential of the current average $\langle I_n(t)\rangle$ at terminal n either with respect to $V_{n'}(t')$ or to $X(t')$ can be expressed through a generalized response formula keeping all V_n and X finite.

connected continuously or by tunneling terms. Then each lead n has a uniform voltage $V_n(t)$. The coupling Hamiltonian reads $\mathcal{H}_1(t) = \int d^d\vec{r}\,V(\vec{r},t)\rho(\vec{r})$, where $\rho(\vec{r})$ is the charge density at space coordinate $\vec{r}$, and d is the dimension. By using the fact that the potential is piecewise constant, this becomes

$$\mathcal{H}_1(t) = \sum_{n=0}^{N} Q_n V_n(t). \tag{4}$$

More generally, even with no recourse to leads, this coupling was adopted, for instance by Büttiker,[8] without a detailed description of the reservoirs.

It is now possible to express in a microscopic way the out-of-equilibrium differential conductance with no constraint either on the Hamiltonian or on the initial density matrix. We use Eq. (1), where $X(t)$ is replaced by $V_n(t)$, upon which the interaction Hamiltonian in Eq. (4) now depends linearly, while there is no explicit dependence of I_n on $V_{n'}$. By denoting $G_{nn'}(t,t') = \delta I_n(t)/\delta V_{n'}(t')$ for $n,n' = 0,\ldots,N$, we get the generalized Kubo formula

$$\hbar G_{nn'}(t,t') = i\theta(t-t')\langle[I_n(t),Q_{n'}(t')]\rangle, \tag{5}$$

where the average is calculated still *in the presence* of the time-dependent Hamiltonian $\mathcal{H}_0(t) + \mathcal{H}_1(t)$, and keeping $V_{n'}(t')$ finite.

We have to stress that the gate is implicitly included in the $N+1$ terminals. This ensures current conservation; see Eq. (3). It turns out that the same conservation law in Eq. (2) guarantees gauge invariance automatically: a translation of all potentials $V_n(t)$ by the same function $V(t)$ has no effect on $\mathcal{H}_1(t)$ in Eq. (4). Let us introduce the double Fourier transform of a function $F(t,t')$ defined as

$$F(\omega,\Omega) = \int\int dx\,ds\,e^{i\omega x + i\Omega s}\tilde{F}(x,s), \tag{6}$$

where we have performed the change of variable $F(t,t') = \tilde{F}(x,s)$ with $s = t+t'$ and $x = t-t'$. By using Eqs. (2), (3), and (5), we get simultaneously the two constraints

$$\sum_{n=0}^{N} G_{nn'}(\omega,\Omega) = \sum_{n'=0}^{N} G_{nn'}(\omega,\Omega) = 0, \tag{7}$$

in which the second one corresponds to gauge invariance, which is tied to the first one. We emphasize that time-reversal symmetry is not necessarily required. Nevertheless, the FF differential conductance matrix $\boldsymbol{G}$ still has some symmetries imposed by the fact that $\delta I_n(t)$ and $\delta V_{n'}(t')$ are real quantities,

$$G^*_{nn'}(\omega,\Omega) = G_{nn'}(-\omega,-\Omega), \tag{8}$$

where the star denotes the complex conjugate. Furthermore, due to causality with respect to $t-t'$, its real and imaginary part obey the Kramers-Krönig relations

$$\mathrm{Im}\,G_{nn'}(\omega,\Omega) = PP\int d\omega' \frac{\mathrm{Re}G_{nn'}(\omega',\Omega)}{\omega-\omega'},$$

where PP denotes the principal part of the integral.

Now we consider the nonsymmetrized current fluctuations matrix $\boldsymbol{S}(t,t')$ whose elements are given by

$$S_{nn'}(t,t') = \langle \hat{I}_{n'}(t')\hat{I}_n(t)\rangle - I_{n'}(t')I_n(t). \tag{9}$$

They obey the symmetry $S_{n'n}(t',t) = S^*_{nn'}(t,t')$, whose Fourier transforms read

$$S_{n'n}(\omega,\Omega) = S^*_{nn'}(\omega,-\Omega). \tag{10}$$

Let us consider

$$\boldsymbol{S}^{\pm}(t,t') = \boldsymbol{S}(t,t') \pm \boldsymbol{S}^T(t',t), \tag{11}$$

which are the symmetric and the antisymmetric parts of $\boldsymbol{S}$ where the subscript T refers to the transpose. In view of Eq. (5), one can show easily that

$$\hbar\partial_{t'}G_{nn'}(t,t') - \hbar\partial_t G_{n'n}(t',t) = iS^-_{nn'}(t,t')$$

(notice that the δ function with respect to $t-t'$ cancels), which, once Fourier transformed, becomes [see Eqs. (9) and (5)]

$$\begin{aligned} S^-_{nn'}(\omega,\Omega) &= S_{nn'}(\omega,\Omega) - S_{n'n}(-\omega,\Omega) \\ &= \hbar(\omega-\Omega)G_{nn'}(\omega,\Omega) + \hbar(\omega+\Omega)G_{n'n}(-\omega,\Omega), \end{aligned} \tag{12}$$

or in the matrix form,

$$\boldsymbol{S}^-(\omega,\Omega) = -\hbar\,(\omega-\Omega)\,\boldsymbol{G}(\omega,\Omega) - \hbar\,(\omega+\Omega)\,\boldsymbol{G}^\dagger(\omega,-\Omega). \tag{13}$$

This equation is the central result of the paper. It is a completely general FDT for the asymmetric part of the matrix $\boldsymbol{S}$. It is valid in an out-of-equilibrium regime and for time-dependent Hamiltonian and voltages, including in the presence of a finite magnetic field that would break time-reversal symmetry.

One consequence of this expression is relating both symmetrized and nonsymmetrized current fluctuations [see Eq. (11)], as one can inject it into the right-hand side (r.h.s.) of

$$2\boldsymbol{S} = \boldsymbol{S}^+ + \boldsymbol{S}^-. \tag{14}$$

If we consider the case $\Omega = 0$ to get the dc component of $\boldsymbol{S}$, then $\boldsymbol{S}(\omega,0)$ and hence $\boldsymbol{S}^{\pm}(\omega,0)$ are now Hermitian, in view of Eq. (10), in particular, their diagonal elements are real. Now Eq. (13) reduces to

$$\boldsymbol{S}^-(\omega,0) = -2\hbar\omega\boldsymbol{G}^h(\omega,0), \tag{15}$$

where the Hermitian part of the matrix $\boldsymbol{G}$ is given by

$$2\boldsymbol{G}^h = \boldsymbol{G} + \boldsymbol{G}^\dagger. \tag{16}$$

Notice that if the potentials in all the terminals are periodic with the same frequency Ω_0, then Ω must be commensurate with Ω_0 [i.e., $\Omega = (l/k)\,\Omega_0$, with l,k integers], but ω can take arbitrary values.

A. Stationary regime

We will focus in the following on both time-independent Hamiltonian and potentials in the reservoirs. Then the time translation invariance is restored [$\boldsymbol{F}(t,t') = \boldsymbol{F}(t-t')$] for $\boldsymbol{F} = \boldsymbol{S},\boldsymbol{G}$, and one requires $\Omega = 0$ in Eqs. (5) and (13). Let us keep similar notations but stress the dependence of $\boldsymbol{F} = \boldsymbol{S},\boldsymbol{G}$ on the voltage vector $\boldsymbol{V} = (V_0,V_1,\ldots,V_N)$ by denoting

$$\boldsymbol{F}(\omega,\Omega) = \delta(\Omega)\boldsymbol{F}(\boldsymbol{V};\omega),$$

where

$$F(V;\omega) = \int dt e^{i\omega(t-t')} F(t-t').$$

For convenience, we also introduce the matrices for the "excess FF differential conductance" and excess current fluctuations:

$$\Delta F(V;\omega) = F(V;\omega) - F(V=0;\omega). \tag{17}$$

As noticed above for the case $\Omega = 0$, the matrices $S(V;\omega), S^{\pm}(V;\omega)$ are Hermitian, while $G(V;\omega)$ is not. Only the diagonal elements of $S(V;\omega)$, which are real, can be referred to as FF noise; $S_{nn}(V;\omega)$ for positive (negative) ω corresponds then to the emitted (absorbed) noise spectrum in terminal n. The asymmetric part of the whole matrix $S(V;\omega)$ follows Eq. (15):

$$S^{-}(V;\omega) = -2\hbar\omega G^{h}(V;\omega). \tag{18}$$

In particular, the diagonal elements read

$$S^{-}_{nn}(V;\omega) = -2\hbar\omega \mathrm{Re} G_{nn}(V;\omega). \tag{19}$$

Equation (18) generalizes, to a much larger extent, that obtained for the scalar FF noise in specific systems in Refs. 29 and 27. It offers an out-of-equilibrium FDT for the asymmetric part of the fluctuation matrix $S(V;\omega)$. Indeed, one can use it to obtain directly the standard equilibrium FDT. At equal voltages in the $N+1$ terminals (taken as zero for convenience), and specifying to a thermal distribution, one has the detailed balance equation $S(V=0;-\omega) = e^{\beta\omega} S(V=0;\omega)$. Thus, Eqs. (18) and (11) yield

$$S(V=0;\omega) = 2\hbar\omega N(\omega) G^{h}(V=0;\omega), \tag{20}$$

where $N(\omega) = 1/(-1+e^{\beta\omega})$. Recall that Eq. (14) establishes the link between the symmetrized and nonsymmetrized current fluctuations. Similarly, the FF fluctuations for negative (respectively positive) frequencies can be deduced from those at positive (respectively negative) frequencies. An interesting alternative is to deduce $G^{h}(V;\omega)$ from $S^{-}(V;\omega)$. This allows one to get rid of any undesirable background sources of noise, which would appear in the same way in both $S(V;\omega)$ and $S(V;-\omega)$, and thus would cancel when taking their difference, $S^{-}(V;\omega)$. Another advantage is that the measurement of the current correlations is not subject to the limitations on frequencies as the FF conductance: these are due to capacitive effects and to the equilibration condition in the reservoirs, $\omega\tau_{in} \ll 1$, where τ_{in} is the inelastic time, in order to define a quasiequilibrium distribution.[22,30,36]

An important feature which Eq. (18) clarifies concerns the asymmetry of $\Delta S(V;\omega)$, in Eq. (17), with respect to positive/negative frequencies. While many theoretical works dealt with the symmetrized noise, it turns out that most experiments are based on quantum detectors measuring the nonsymmetrized excess noise,[37,38] which has been the subject of few theoretical works dealing with electronic interactions.[29–31,39,40] In a coherent conductor with energy-independent transmission, the excess noise is identical whether or not one symmetrizes the current-current correlator, i.e., $\Delta S^{+}(V;\omega) = \Delta S(V;\omega)$, which is therefore symmetric with respect to positive/negative frequencies.[11] An important question is which criterion could violate the symmetry obtained there, thus giving evidence for a quantum measurement. It is interesting to discuss the asymmetry of the matrix $\Delta S(V;\omega)$ within our formalism, and thus to find the criterion for asymmetric, excess cross correlations as well. This can be achieved by using Eq. (18) [see Eqs. (11) and (17)]:[41]

$$\Delta S(V;-\omega) - \Delta S^{T}(V;\omega) = 2\hbar\omega \Delta G^{h}(V;\omega).$$

Therefore, the asymmetry between $\Delta S_{nn'}(V;-\omega)$ and $\Delta S_{n'n}(V;\omega)$ requires that $\Delta G^{h}_{nn'}(V;\omega) \neq 0$. This yields a necessary criterion, i.e., a nonlinear regime, in the sense that the FF differential conductance depends on the dc voltage. However, this criterion is not sufficient for different terminals $n \neq n'$: one could have $G^{h}_{nn'}(V;\omega) = 0$ at any V, which ensures instead symmetry of both cross correlations and their excess value, even in the nonlinear regime. This feature has been obtained in chiral edges of the FQHE:[40] in the corresponding setup, some nondiagonal elements of the conductance matrix vanish due to chirality. At the same time, excess autocorrelations were found to be asymmetric in that setup, as well as in quantum wires and carbon nanotubes,[30] due to the nonlinear behavior associated with backscattering in the presence of interactions. Obviously, interactions are not necessary to induce nonlinearity. The FF noise has been, for instance, studied in noninteracting systems where the transmission is energy dependent, such as a wire with two barriers,[42] hybrid structures,[43] and Josephson junctions where the asymmetry between the emission and absorption of the excess FF noise has been measured experimentally.[38] Recently, the FF noise through a quantum dot has been investigated,[31] and the dissipative nonlinear FF conductance expressed using Eq. (18). Thus nonlinearity with respect to the dc voltage is a common origin for the asymmetry obtained in all those various systems.

Turning back to an arbitrary, multiterminal, nonlinear system, it is instructive to introduce a combination we call a "modified" excess current fluctuations matrix:

$$\check{\Delta} S(V;\omega) = S(V;\omega) - 2\hbar\omega N(\omega) G^{h}(V;\omega). \tag{21}$$

The second term on the r.h.s. of Eq. (21) is inspired by Eq. (20), and would be identical to equilibrium fluctuations in a linear system, where $\check{\Delta} S(V;\omega) = \Delta S(V;\omega)$ becomes identical to the excess current fluctuations matrix. Indeed, such an identity holds in a nonlinear system in the quantum regime, i.e., for positive frequencies obeying $\hbar\omega \gg k_B T$. For arbitrary frequencies, $\check{\Delta} S(V;\omega)$ has analogous properties to those of $\Delta S(V;\omega)$ in linear systems: it vanishes at equilibrium ($\check{\Delta} S(V=0;\omega) = 0$ [Eq. (20)]), and more interestingly, it restores the symmetry ($\check{\Delta} S(V;-\omega) = \check{\Delta} S(V;+\omega)$).

In view of Eq. (18), we can also express it as

$$\check{\Delta} S(V;\omega) = [1+N(\omega)] S(V;\omega) - N(\omega) S(V;-\omega). \tag{22}$$

Notice that if one replaces the electron temperature T in $N(\omega)$ by that of a detector, the r.h.s. would be the quantity expected to be measured in a linear system, as predicted by Lesovik and Loosen in the case of a scalar noise.[27]

Now we show how the out-of-equilibrium FDT, given by Eq. (18), solves the paradox of the negative sign of the excess noise. This looks counterintuitive, since by applying a bias, we expect to induce more noise (hence the qualifier "excess").[44]

We focus on autocorrelations as they are real and can be interpreted in terms of the emission and absorption spectrum. In two-terminal geometries, they were found to be negative for certain frequencies, such as in TLLs, whether symmetrization is performed[45,46] or not,[30,40] and without interactions for an energy-dependent transmission.[43,45] Indeed, for $\hbar\omega \gg k_B T$, one has

$$\Delta S_{nn}(\boldsymbol{V};\omega) = S_{nn}(\boldsymbol{V};\omega), \tag{23}$$

which is the correlator of the same current at terminal n, and thus (using a spectral decomposition) is always positive. Therefore, $\Delta S_{nn}(\omega \gg T) \geqslant 0$. Nevertheless, the absorption excess noise $\Delta S_{nn}[-(\omega \gg T)]$ can be negative for frequencies and voltages where $\mathrm{Re}\Delta G_{nn}(\boldsymbol{V};\omega)$ is negative and large enough; see Eqs. (17) and (19). Finally, the symmetrized excess noise $\Delta S^+_{nn}(\boldsymbol{V};\omega) = \Delta S_{nn}(\boldsymbol{V};\omega) + \Delta S_{nn}(\boldsymbol{V};-\omega)$, being a superposition of both emission and absorption, can be negative too.

IV. APPLICATION TO A TUNNEL JUNCTION

Let us now consider a tunnel junction between two conductors with arbitrary dimensions, possibly with disorder and internal or mutual interactions encoded in a Hamiltonian $\mathcal{H}_0$. We could allow for many tunneling processes transferring different charges, but for purely pedagogical reasons, we consider a unique process transferring a charge q. Thus the starting model is similar to that in Ref. 47 (see Fig. 2), but with no restrictions on the supercurrent. We require only to have well-defined charges for both systems Q_1 and Q_2, which are conserved by $\mathcal{H}_0$, i.e., that $[Q_l,\mathcal{H}_0] = 0$ for $l = 1,2$. We allow for possible explicit time dependence of the tunneling operators $\mathcal{T}(t)$ (in the Schrödinger representation) and of which we do not specify the form. Indeed, we allow for the tunneling amplitudes to depend on both right- and left-side states, as well as on time. But, contrary to Ref. 47, it is not necessary for our present purpose to require the same time dependence for different states. Finally, we allow for a magnetic field breaking the time-reversal symmetry, and coupling to an arbitrary electromagnetic environment could be incorporated. As we do not need to specify the density matrix, one could deal with many subsystems with different distributions, for instance, the case where the tunnel junction has a thermal distribution with a temperature different from that of the environment. Both conductors are subject to time-dependent potentials $V_1(t)$, $V_2(t)$, and we denote $V(t) = V_1(t) - V_2(t)$. We introduce $\dot{\phi}(t) = V(t)/\hbar$. The coupling of the tunneling charges to these potentials can be absorbed by a gauge transformation. Here we skip the details, described in Ref. 47, and write the total Hamiltonian in its final form:

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_T(t),$$
$$\mathcal{H}_T(t) = e^{iq\phi(t)}\mathcal{T}(t) + e^{-iq\phi(t)}\mathcal{T}^\dagger(t). \tag{24}$$

Thus the tunneling current operator across the tunnel junction is given by

$$\hat{I}(t) = -i\frac{q}{\hbar}[e^{iq\phi(t)}\mathcal{T}(t) - e^{-iq\phi(t)}\mathcal{T}^\dagger(t)]. \tag{25}$$

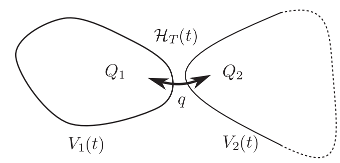

FIG. 2. Tunnel junction between two conductors 1 and 2 for arbitrary dimension, interactions or disorder, subject to time-dependent potential $V_{1,2}(t)$, and with time-dependent tunneling (not necessarily weak) of charges q. These conductors can be either similar or different, such as fractional edge states with equal or different filling factors, leads that are superconducting, insulating, or normal, etc. One can also view one system as a probe, such as the tip of a scanning tunneling microscope or a Fermi liquid coupled to edge states or to a quantum dot. Capacitive coupling is possible, and we require only charges Q_1 and Q_2 to be well defined, commuting with the total Hamiltonian. Coupling to an electromagnetic environment can be incorporated as well.

A. Generalized Kubo formula and FDT

Our goal is to express the associated nonequilibrium, time-dependent conductance

$$G(t,t') = \frac{\delta I(t)}{\delta V(t')},$$

where the average tunneling current is

$$I(t) = \langle \hat{I}(t)\rangle.$$

Using (1) and the fact that $\partial\hat{I}/\partial\phi = \frac{q^2}{\hbar}\mathcal{H}_T(t)$, we can derive the nonperturbative expression

$$\partial_{t'}G(t,t') = \chi(t,t') - \chi_0\left(t,t'\right), \tag{26}$$

where

$$\chi(t,t') = \frac{i}{\hbar}\theta(t-t')\,\langle\,[\hat{I}(t),\hat{I}(t')]\,\rangle\,,$$

and

$$\chi_0\left(t,t'\right) = \delta\left(t-t'\right)\left(\frac{q}{\hbar}\right)^2\langle\mathcal{H}_T\left(t\right)\rangle\,.$$

This exact Kubo-like expression of the differential conductance $G(t,t')$ is different from known results in that the averages taken here are still in the presence of an arbitrary time dependent finite voltage $V(t)$ and tunneling amplitudes. Note also that in addition to the current commutator, one has a nontrivial analogous of a diamagnetic term (χ_0) obtained usually in the conductivity.

If one assumes now the absence of any Josephson current, one can use the gauge invariance to show that $\chi_0(t,t') = \delta(t-t')\int dt''\chi(t,t'')$, which in the frequency domain yields $\chi_0(\omega,\Omega) = \chi(\Omega,\Omega)$, independent of ω due to the δ function with respect to $t-t'$. Then, the Fourier transform Eq. (7) of $G(t,t')$ reads

$$i(\omega-\Omega)G(\omega,\Omega) = \chi(\omega,\Omega) - \chi(\Omega,\Omega).$$

Note that the out-of-equilibrium FDT for the asymmetric part of the current fluctuations in Eq. (18) is not affected by the χ_0 term,

$$\begin{aligned}S^-(t,t') &= S(t,t') - S(t',t)\\ &= \partial_{t'}G(t,t') - \partial_t G(t,t'),\end{aligned} \tag{27}$$

where the nonsymmetrized current correlator [as in Eq. (9)] is

$$S(t,t') = \langle \hat{I}(t')\hat{I}(t)\rangle - I(t')I(t). \tag{28}$$

The exact relation (26) was shown to hold in the limit of weak tunneling in Ref. 47, while (27) will be shown to hold in the same limit in a separate paper.[48] More interestingly, the last work will include other universal FDT relations as FF current fluctuations and the FF differential conductance can be expressed in terms of the dc current. The remainder of the present paper we also focus on this weak tunneling limit.

B. Finite frequency noise in the weak tunneling limit

Being related to the asymmetry discussed in the above, we choose to present here a universal property of the FF noise, $S(V;\omega) = \int dt S(t,t')e^{i\omega(t-t')}$, in the stationary regime where $S(t,t') = S(t-t')$ (implying $\Omega = 0$), i.e., when both the tunneling amplitudes and the voltage are time independent, and thus $\mathcal{T}(t) = \mathcal{T}$ and $V(t) = V$. We further assume that tunneling is weak so that we can use the equilibrium density matrix of the unperturbed system, and, in addition, that $\langle \mathcal{T}\mathcal{T}\rangle = 0$, thus a possible supercurrent has to be negligible. Then, to the lowest order in tunneling, one can simply express the FF noise given by Eq. (28) through a spectral decomposition over the many-body exact eigenstates $|\alpha\rangle$ of the Hamiltonian $\mathcal{H}_0$ [see Eq. (24)]:

$$S(V;\omega) = 2\pi q^2 \sum_{\alpha,\alpha'} [\rho_\alpha \delta(\hbar\omega - qV + E_{\alpha'} - E_\alpha) + \rho_{\alpha'}\delta(\hbar\omega + qV - E_{\alpha'} + E_\alpha)]\,|\langle\alpha\,|\mathcal{T}(0)|\alpha'\rangle|^2,$$

where $\rho_\alpha = \langle\alpha\,|\rho|\,\alpha\rangle$ is the diagonal element of the equilibrium density matrix. If we assume it to be thermal, i.e., $\rho = e^{-\beta\mathcal{H}_0}/Z$, one has $\rho_\alpha = e^{-\beta(E_\alpha - E_0)}\rho_0$, where 0 labels the ground state (or one of the ground states in case they are degenerate), and $\beta = (k_B T)^{-1}$. Note that since one can exchange the indices α and α', $S(V;\omega)$ is even in V and can be expressed as

$$\begin{aligned} S(V;\omega) = 2\pi q^2 \rho_0 \sum_{\alpha,\alpha'} [&e^{-\beta(-|qV| + \hbar\omega + E_{\alpha'} - E_0)} \\ &\times \delta(\hbar\omega - qV + E_{\alpha'} - E_\alpha) \\ &+ e^{-\beta(|qV| + \hbar\omega + E_\alpha - E_0)}\delta(\hbar\omega + qV - E_{\alpha'} + E_\alpha)] \\ &\times |\langle\alpha|\mathcal{T}(0)|\alpha'\rangle|^2, \end{aligned}$$

where the differences $E_{\alpha'} - E_0$ and $E_\alpha - E_0$ are positive. Let's consider ω positive. Then, as soon as

$$\hbar\omega - |qV| \gg k_B T, \tag{29}$$

all exponentials vanish. This shows a universal property of the emitted noise, which is independent on the quantum many-body states in both electrodes: it vanishes at frequencies obeying Eq. (29). We do not expect this result to be valid to higher orders in tunneling, being related to energy conservation of one quasiparticle tunneling. In a future paper,[48] we will show a universal FDT-type expression of the nonsymmetrized noise $S(V;\omega)$ in terms of the dc current only, which allows one to prove the same property as well. For frequencies obeying Eq. (29), using Eq. (19) yields in contrast a non-vanishing absorption noise in that frequency range:

$$S(V;-\omega) = 2\hbar\omega \mathrm{Re}\, G(V;\omega),$$

where $G(V;\omega) = \int dt G(t,t')e^{i\omega(t-t')}$, and G is expressed in Eq. (26). This later equation expresses a generalized out-of-equilibrium FDT for the absorption noise in the quantum regime. Note that in that regime the absorption noise is not necessarily linear in omega. We now consider the excess noise, still in the frequency range defined by Eq. (29). In that range Eq. (23) is valid and thus the excess emitted noise vanishes as the emitted noise, while the excess absorption noise is given by:

$$\Delta S(V;-\omega) = 2\hbar\omega[\mathrm{Re}\, G(V;\omega) - \mathrm{Re}\, G(0;\omega)].$$

In particular, in the limit of zero temperature, Eq. (29) becomes $\hbar\omega > |qV|$, from which we conclude that the excess FF noise vanishes for $\hbar\omega > |qV|$ (emitted noise) but not for $-\hbar\omega < -|qV|$ (absorption noise) for systems that are non-linear with respect to the dc voltage V.

Consequently, the symmetrized excess noise, being the mean of emission and absorption spectrum, does not vanish either for $|\hbar\omega| > |qV|$ in nonlinear systems. This gives a common explanation for the behavior of the FF noise obtained in the case of one-dimensional systems with weak or strong backscatttering.[30,40,45,46]

We have to draw attention to the important facts that matter if one-dimensional systems are to be considered. First, the above generalized Kubo formula can be applied not only for tunneling barriers, but also for arbitrary backscattering, even spatially extended. Nevertheless, in both limits, the total current operator in the electrodes is, strictly speaking, different from the tunneling or backscattering current operator. When its average is measured at the junction, but at some distance L, the total current, conductance, and current fluctuations coincide with their values given in expressions (25), (26), and (28) only for low enough frequencies $\omega L/v \ll 1$, with v being a typical velocity, e.g., that of plasmonic modes. The difference was explicitly established in Refs. 30, 40, and 45, and is partly due to the propagation of plasmons from the tunneling junction or backscattering centers along the distance L. At higher frequencies, one has to use instead the expression for the conductance in Eq. (5), where the current operator is that defined at the contacts. We do not expect such a difference if the electrodes are two or three dimensional, as oscillating contributions due to the propagation in the electrodes are expected to be washed out.

V. CONCLUSIONS

To conclude, we have derived a general time-dependent response formula for a Hamiltonian depending in an arbitrary way on time-dependent parameters [Eq. (1)]. When applied to a multiterminal mesoscopic system described by a time-dependent Hamiltonian, this formula yields a microscopic and current-conserving expression of the differential conductance matrix [Eq. (5)]. Its elements consist of the derivative of the average current at a given terminal and time with respect to the potential at any other terminal and time. We thus provide a promising framework to study systematically time-dependent transport in nonlinear systems, including strongly correlated ones. This formulation is expected to be useful for investigating pumping or mixing setups. It is noteworthy that we are able to express higher-order derivatives of any operator average. Thus we can express the variation of the current in one terminal in response to change in the potentials in many terminals.[49]

We have used this framework to derive a universal time-dependent, out-of-equilibrium fluctuation-dissipation theorem (FDT) for the asymmetric part of the current fluctuations matrix [Eq. (12)]. In the stationary regime, the FDT applied to current autocorrelations states that the difference between the emitted and absorption spectrum in terminal n is related to the dissipation in an out-of-equilibrium situation. We extend the FDT to cross correlations, provided the appropriate combination of nondiagonal elements of the conductance matrix are taken into account. We show that the FDT sheds light on the asymmetry of the excess FF current fluctuations matrix, and on the negative sign of its diagonal elements in certain systems for some frequencies and voltages. We show that nonlinearity is a common origin for both features, which explains their occurrence in various nonlinear systems studied previously. Furthermore, we propose a "modified" FF excess noise showing properties that are analogous to the excess fluctuation matrix in linear systems. In particular, it restores symmetry with respect to positive and negative frequencies.

Then, we have explicited these time-dependent Kubo formula and FDT in the case of a nonlinear tunnel junction in the presence of arbitrary voltage, interactions within or between the electrodes (even extended ones), tunneling strength, and time dependence. Finally, restricting to the weak tunneling limit (or weak backscattering in 1D) and negligible supercurrent, we have derived a universal property of the FF emitted noise.

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