Current Noise in ac-Driven Nanoscale Conductors

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The theory for current fluctuations in ac-driven transport through nanoscale systems is put forward. By use of a generalized, non-Hermitian Floquet theory we derive novel explicit expressions for the time-averaged current and the zero-frequency component of the power spectrum of current fluctuations. A distinct suppression of both the zero-frequency noise and the dc current occurs for suitably tailored ac fields. The relative level of transport noise, being characterized by a Fano factor, can selectively be manipulated by ac sources; in particular, it exhibits both characteristic maxima and minima near current suppression.

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Recent experimental successes in the coherent coupling of quantum dots [1] and in the reproducible measurement of electronic currents through molecules [2,3] have given rise to renewed theoretical interest in the transport properties of nanoscale systems [4,5]. Thereby, new ideas in order to exploit the quantum coherence of such systems for the construction of novel electronic devices [5] have emerged. One possible construction element is based on the manipulation of quantum dots or single molecules by use of an oscillating gate voltage or an infrared laser, respectively. A prominent effect of such ac fields consists of the adiabatic [6–9] and nonadiabatic [10,11] pumping of electrons. Moreover, laser irradiated molecular wires provide novel devices such as coherent quantum rectifiers [12] and optically controlled transistors [13]. However, such time-dependent control schemes can be valuable in practice only if they operate at tolerable noise levels. Thus, the question whether noise properties of nanoscale systems can be selectively manipulated becomes of foremost interest.

Electron transport through time-independent, mesoscopic systems is commonly described within the framework of a scattering formalism. Both the average current [14] and the transport noise characteristics [15,16] can be expressed in terms of the quantum transmission coefficients for the corresponding transport channels. By contrast, the theory for driven quantum transport is much less developed. Expressions for the spectral density of the current fluctuations have been derived for the low-frequency ac conductance [17] and the scattering by a slowly time-dependent potential [18]. However, the situation becomes more opaque in the presence of rapidly varying time-dependent fields. Within a Green function approach, a *formal* expression for the current through a time-dependent conductor has been presented in Refs. [19,20]. Here, we derive *explicit* expressions for both the current and the noise properties of electron transport through a nanoscale conductor under the influence of time-dependent forces at arbitrary frequency and strength. The dynamics of the electrons is solved by integrating the Heisenberg equations of motion for the electron creation/annihilation operators within a generalized Floquet approach. We then use the resulting expressions to explore the possibility of an *a priori* control of the dc current and the zero-frequency noise by the influence of an ac field.

The lead-wire model.—The entire setup of our nanoscale system is described by the time-dependent Hamiltonian $H(t) = H_{\mathrm{wire}}(t) + H_{\mathrm{leads}} + H_{\mathrm{contacts}}$, where the different terms correspond to the driven wire (or coupled quantum dots), the leads, and the wire-leads coupling, respectively. In order to go beyond merely formal considerations, we herewith focus on the regime of *coherent quantum transport* where the main physics at work occurs on the wire itself. In doing so, we neglect other possible influences stemming from driving induced hot electrons in the leads, dissipation on the wire, and, as well, electron-electron interaction effects. Then, the wire Hamiltonian reads in a tight-binding approximation with N orbitals $|n\rangle$

$$H_{\mathrm{wire}}(t) = \sum_{n,n'} H_{nn'}(t) c_n^\dagger c_{n'}. \tag{1}$$

The fermion operators c_n and $c_n^\dagger$ annihilate and create, respectively, an electron in the orbital $|n\rangle$. The influence of an applied ac field with frequency $\Omega = 2\pi/\mathcal{T}$ results in a periodic time dependence of the Hamiltonian: $H_{nn'}(t+\mathcal{T}) = H_{nn'}(t)$. The leads are modeled by ideal electron gases, $H_{\mathrm{leads}} = \sum_q \epsilon_q (c_{Lq}^\dagger c_{Lq} + c_{Rq}^\dagger c_{Rq})$, where $c_{Lq}^\dagger$ ($c_{Rq}^\dagger$) creates an electron in the state $|Lq\rangle$ ($|Rq\rangle$) in the left (right) lead. The tunneling Hamiltonian

$$H_{\mathrm{contacts}} = \sum_q (V_{Lq} c_{Lq}^\dagger c_1 + V_{Rq} c_{Rq}^\dagger c_N) + \mathrm{H.c.} \tag{2}$$

establishes the contact between the sites $|1\rangle$, $|N\rangle$, and the respective lead, as sketched in Fig. 1. Below, we shall assume within a so-termed wide-band limit that the coupling strengths $\Gamma_\ell = 2\pi \sum_q |V_{\ell q}|^2 \delta(\epsilon - \epsilon_q)$, $\ell = L, R$ are energy independent. To specify fully the dynamics,

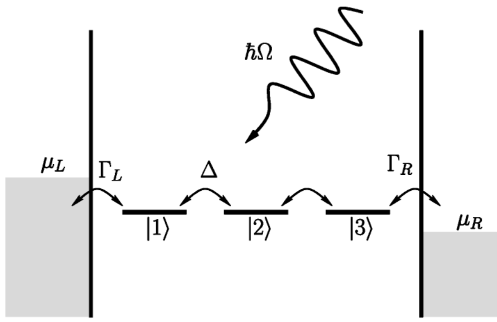

FIG. 1. Level structure of the molecular wire with $N = 3$ orbitals. The end sites are coupled to two leads with chemical potentials μ_L and $\mu_R = \mu_L - eV$.

we choose as an initial condition for the left/right lead a grand-canonical electron ensemble at temperature T and electrochemical potential $\mu_{L/R}$, respectively. An applied voltage V maps to a chemical potential difference $\mu_R - \mu_L = eV$, where $-e$ is the electron charge.

We shall focus on two central transport quantities: the time-dependent electrical currents through the two contacts and their fluctuations. The current operators are given by the negative time variation of the electron numbers in the leads, multiplied by the electron charge $-e$, $I_\ell(t) = ie[H(t), N_\ell]/\hbar$, where $N_\ell = \sum_q c^\dagger_{\ell q} c_{\ell q}$ denotes the electron number in lead ℓ.

Generalized Floquet approach.—For the evaluation of correlation functions, we work in the Heisenberg picture and derive the annihilation operators at long times by a Floquet ansatz [21]. From the Hamiltonian $H(t)$ follow the Heisenberg equations for the lead operators, with the wire operators appearing in an inhomogeneity. In an integrated form they read

$$c_{Lq}(t) = c_{Lq}(t_0)e^{-i\epsilon_q(t-t_0)/\hbar} - \frac{iV_{Lq}}{\hbar}\int_0^{t-t_0} d\tau e^{-i\epsilon_q\tau/\hbar} c_1(t-\tau)$$

and $c_{Rq}(t)$ accordingly. Inserting this into the Heisenberg equations for the wire operators yields

$$\begin{aligned} \dot{c}_{1/N} &= -\frac{i}{\hbar}\sum_{n'} H_{1/N,n'}(t)c_{n'} - \frac{\Gamma_{L/R}}{2\hbar}c_{1/N} + \xi_{L/R}(t), \\ \dot{c}_n &= -\frac{i}{\hbar}\sum_{n'} H_{nn'}(t)c_{n'}, \qquad n = 2, \ldots, N-1. \end{aligned} \tag{3}$$

Owing to the wide-band limit, the dissipative terms are memory free. Within the chosen grand-canonical ensembles the operator-valued Gaussian noise $\xi_\ell(t) = -(i/\hbar)\sum_q V^*_{\ell q} e^{-i\epsilon_q(t-t_0)/\hbar} c_{\ell q}(t_0)$ obeys

$$\langle \xi_\ell(t) \rangle = 0, \tag{4}$$

$$\langle \xi^\dagger_\ell(t)\xi_{\ell'}(t')\rangle = \delta_{\ell\ell'}\frac{\Gamma_\ell}{2\pi\hbar^2}\int d\epsilon e^{i\epsilon(t-t')/\hbar} f_\ell(\epsilon), \tag{5}$$

where $f_\ell(\epsilon) = (1 + \exp[(\epsilon - \mu_\ell)/k_B T])^{-1}$ denotes the Fermi function at temperature T and chemical potential μ_ℓ, $\ell = L, R$. The current operator then assumes the form

$$I_L(t) = \frac{e}{\hbar}\Gamma_L c^\dagger_1(t)c_1(t) - e\{c^\dagger_1(t)\xi_L(t) + \xi^\dagger_L(t)c_1(t)\}. \tag{6}$$

Before solving the inhomogeneous set of Eqs. (3), let us first analyze the corresponding homogeneous equations. They are linear and possess time-dependent, $\mathcal{T}$-periodic coefficients. Thus, it is possible to construct a complete solution with the help of the Floquet ansatz $|\psi_\alpha(t)\rangle = \exp[(-i\epsilon_\alpha/\hbar - \gamma_\alpha)t]|u_\alpha(t)\rangle$. The Floquet states $|u_\alpha(t)\rangle = \sum_k |u_{\alpha k}\rangle \exp(-ik\Omega t)$ obey the time periodicity of the differential equations and fulfill in a Hilbert space that is extended by a periodic time coordinate the eigenvalue equation

$$\left(\mathcal{H}(t) - i\Sigma - i\hbar\frac{d}{dt}\right)|u_\alpha(t)\rangle = (\epsilon_\alpha - i\hbar\gamma_\alpha)|u_\alpha(t)\rangle, \tag{7}$$

where $\mathcal{H}(t) = \sum_{n,n'}|n\rangle H_{nn'}(t)\langle n'|$ and $2\Sigma = |1\rangle\Gamma_L\langle 1| + |N\rangle\Gamma_R\langle N|$. Because the eigenvalue equation (7) is non-Hermitian, its eigenvalues $\epsilon_\alpha - i\hbar\gamma_\alpha$ are generally complex valued and the (right) eigenvectors are not mutually orthogonal. Therefore, we need to solve also the adjoint Floquet equation yielding again the same eigenvalues but providing the adjoint eigenvectors $|u^+_\alpha(t)\rangle$. It can be shown that the Floquet states $|u_\alpha(t)\rangle$ together with the adjoint states $|u^+_\alpha(t)\rangle$ form at equal times a complete biorthogonal basis: $\langle u^+_\alpha(t)|u_\beta(t)\rangle = \delta_{\alpha\beta}$ and $\sum_\alpha |u_\alpha(t)\rangle\langle u^+_\alpha(t)| = \mathbf{1}$ [22]. For $\Gamma_{L/R} = 0$, both $|u_\alpha(t)\rangle$ and $|u^+_\alpha(t)\rangle$ reduce to the usual Floquet states.

The Floquet states $|u_\alpha(t)\rangle$ allow one to write the general solution of Eq. (3) in closed form. In the asymptotic limit $t_0 \to -\infty$, it reads

$$c_n(t) = \sum_\alpha \int_0^\infty d\tau \langle n|u_\alpha(t)\rangle e^{(-i\epsilon_\alpha/\hbar - \gamma_\alpha)\tau}\langle u^+_\alpha(t-\tau)| \times \{|1\rangle\xi_L(t-\tau) + |N\rangle\xi_R(t-\tau)\}. \tag{8}$$

To obtain the current $\langle I_L(t)\rangle$, we insert the operator (8) into Eq. (6) and use the expectation values (5). We then find that the current assumes the commonly expected "scattering form" [14] but with *time-dependent* transmission probabilities and, as well, an additional contribution that accounts for a $\mathcal{T}$-periodic charging of the wire. The latter does not contribute to the time-averaged current $\bar{I} = \int_0^{\mathcal{T}} dt\langle I_L(t)\rangle/\mathcal{T}$ so that we obtain as a first result

$$\bar{I} = \frac{e}{2\pi\hbar}\sum_{k=-\infty}^{\infty}\int d\epsilon\{T^{(k)}_{LR}(\epsilon)f_R(\epsilon) - T^{(k)}_{RL}(\epsilon)f_L(\epsilon)\}. \tag{9}$$

Owing to charge conservation it equals the average current through the right contact. The coefficients

$$T^{(k)}_{LR}(\epsilon) = \Gamma_L\Gamma_R|G^{(k)}_{1N}(\epsilon)|^2 \tag{10}$$

can be interpreted as the probability that an electron with energy ϵ is transmitted from the right lead to the left lead under the absorption of k photons, respectively, the emission of $-k$ photons when $k < 0$. Note that the sum runs over all integers k corresponding to different conduction channels that contribute independently to the average current $\bar{I}$. The retarded Green function

$$G_{nn'}^{(k)}(\epsilon) = \sum_{\alpha,k'} \frac{\langle n|u_{\alpha,k'+k}\rangle\langle u_{\alpha,k'}^{+}|n'\rangle}{\epsilon - (\epsilon_\alpha + k'\hbar\Omega - i\hbar\gamma_\alpha)} \tag{11}$$

describes the propagation of an electron from orbital $|n'\rangle$ to orbital $|n\rangle$. We emphasize that generally $|G_{1N}^{(k)}(\epsilon)| \neq |G_{N1}^{(k)}(\epsilon)|$ for a driven system. An expression for the time-averaged current similar to Eq. (9) has been proposed in Ref. [19] without providing an explicit form for the Green function in terms of generalized Floquet states.

Next, we address the main topic of this work, namely, the current noise given by the autocorrelation function $S_L(t,\tau) = \frac{1}{2}\langle[\Delta I_L(t), \Delta I_L(t+\tau)]_+\rangle$ of the current fluctuation operator $\Delta I_L(t) = I_L(t) - \langle I_L(t)\rangle$. It can be shown that $S_L(t,\tau) = S_L(t+\mathcal{T},\tau)$ shares the time periodicity of the driving. Therefore, it is possible to characterize the noise level by the time-averaged noise strength at zero frequency, $\bar{S}_L = \int d\tau \int_0^{\mathcal{T}} dt S_L(t,\tau)/\mathcal{T}$. Since the total charge is conserved, the zero-frequency noise is identical at both the left and the right contact, i.e., $\bar{S}_L = \bar{S}_R = \bar{S}$. After some extensive algebra we obtain our central result

$$\begin{aligned}\bar{S} = \frac{e^2}{2\pi\hbar}\Gamma_L\Gamma_R\sum_k \int d\epsilon\Big\{&\Gamma_L\Gamma_R\Big|\sum_{k'} G_{N1}^{(k'-k)}(\epsilon + k\hbar\Omega)G_{N1}^{(k')}(\epsilon)^*\Big|^2 f_L(\epsilon)\bar{f}_L(\epsilon + k\hbar\Omega)\\ &+ \Big|G_{1N}^{(-k)}(\epsilon + k\hbar\Omega) + i\Gamma_L\sum_{k'} G_{1N}^{(k'-k)}(\epsilon + k\hbar\Omega)G_{11}^{(k')}(\epsilon)^*\Big|^2 f_L(\epsilon)\bar{f}_R(\epsilon + k\hbar\Omega)\Big\}\\ &+ \text{ same terms with the replacement } (L,1) \leftrightarrow (R,N),\end{aligned} \tag{12}$$

where we have defined $\bar{f}_{L/R} = 1 - f_{L/R}$. The key results (9) and (12) contain as special cases prior findings: In the absence of any driving, the Floquet eigenvalues $\epsilon_\alpha - i\hbar\gamma_\alpha$ reduce to the complex-valued eigenenergies; this implies $G_{nn'}^{(k)} = 0$ for all $k \neq 0$, yielding the transmission probability for an electron with energy E of $T(E) = \Gamma_L\Gamma_R|G_{N1}^{(0)}(E)|^2$. Thus, the quantities $\bar{I}$ and $\bar{S}$ agree with the well-known expressions obtained within the time-independent, nondriven scattering approach [15]. For a system for which the ac potential is spatially uniform in the driven region, the average current and the noise strength follow in the low tunneling limit already from the static conduction properties [24]. In the limit of a weak system-lead coupling but arbitrary driving strength, the average current (9) coincides also with the corresponding result of a recent master equation approach [12].

Current and noise suppression.—As a simple, yet nontrivial application, we consider a wire composed of N orbitals as sketched in Fig. 1. Each orbital is coupled to its nearest neighbors by a hopping matrix element Δ. The on-site energies are modulated by the influence of the ac dipole field, $H_{nn}(t) = E_n - A\cos(\Omega t)(N + 1 - 2n)/2$, $n = 1, \ldots, N$. The energy A equals the electric field strength multiplied by the electron charge $-e$ and the distance between two neighboring sites. The wire is assumed to couple equally to both leads, $\Gamma_L = \Gamma_R = \Gamma$, and the laser frequency is far off resonance, $\Omega = 5\Delta/\hbar$. For a molecular wire, a typical value for the hopping matrix element Δ and the coupling strength Γ is 0.1 eV leading to a current unit $e\Gamma/\hbar \simeq 25\ \mu$A, while the laser frequency lies in the optical regime. For a distance of 2 Å between two neighboring sites, a driving amplitude $A = \Delta$ corresponds to an electric field strength of roughly 5×10^6 V/cm. For the evaluation of the current $\bar{I}$ and the noise $\bar{S}$, we restrict ourselves to zero temperature. Then, the Fermi functions turn into step functions and the energy integrals in Eqs. (9) and (12) can be carried out analytically. This limit is physically well justified for molecular wires at room temperature and for quantum dots at helium temperature since in both cases, thermal electron excitations do not play a significant role.

Figure 2(a) depicts the dc current and the zero-frequency noise for a wire with $N = 3$ sites with equal on-site energies and a relatively large applied voltage. As a remarkable feature, we find that for certain values of the field amplitude A, the current drops to a value of some percent of the current in the absence of the field [13]. A perturbation theory for the Floquet equation (7) for $\Delta, \Gamma \ll \hbar\Omega$ yields that the driving results in a renormalized hopping matrix element $\Delta \to \Delta_{\rm eff} = J_0(A/\hbar\Omega)\Delta$, where J_0 denotes the zeroth-order Bessel function. Then, G_{1N} and G_{N1} vanish if the condition $J_0(A/\hbar\Omega) = 0$ is fulfilled [25]. Consequently, the dc current (9) and the zero-frequency noise (12) also vanish.

The *relative* noise strength can be characterized by the so-called Fano factor $F = \bar{S}/e|\bar{I}|$ depicted in Fig. 2(b). Interestingly enough, we find that the Fano factor exhibits as a function of the driving amplitude A both a sharp maximum at current suppression and two pronounced minima nearby. For a sufficiently large voltage, the Fano factor assumes at the maximum the value $F \approx 1/2$. Once the driving amplitude is of the order of the applied voltage, however, the Fano factor becomes much larger. The relative noise minima are distinct and

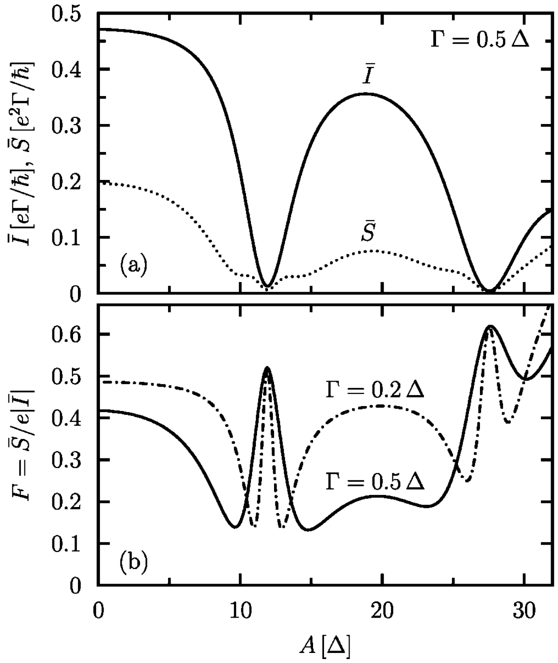

FIG. 2. Time-averaged current $\bar{I}$ and zero-frequency noise $\bar{S}$ (a) as a function of the driving amplitude A for a wire with $N = 3$ sites with on-site energies $E_n = 0$ and chemical potentials $\mu_R = -\mu_L = 25\Delta$. The other parameters read $\Omega = 5\Delta/\hbar$ and $\Gamma = 0.5\Delta$. (b) displays the Fano factor F for these parameters (solid line) and for smaller wire-lead coupling (dash-dotted line).

provide a typical Fano factor of $F \approx 0.15$. Reducing the coupling to the leads renders these phenomena even more pronounced since then the suppressions occur in a smaller interval of the driving amplitude; cf. Fig. 2(b). The overall behavior is robust in the sense that approximately the same values for the minima and the maximum are also found for larger wires, different driving frequencies, different coupling strengths, and slightly modified on-site energies, provided that $\Delta, \Gamma, E_n \ll \hbar\Omega$ and that the applied voltage is sufficiently large. The qualitative behavior can again be understood within a perturbative approach. With increasing driving amplitude, a crossover from $\Delta_{\rm eff} \gg \Gamma$ to $\Delta_{\rm eff} \ll \Gamma$ at the current suppression occurs. Both limits correspond to the transport through a symmetric double barrier and, therefore, are characterized by $F \approx 1/2$ [15]. At the crossover $\Delta_{\rm eff} \approx \Gamma$ the effective barriers vanish and, consequently, the Fano factor assumes its minimum.

In summary, we have put forward with Eqs. (9) and (12) new and appealing expressions for the time-averaged current and the zero-frequency noise for the electron transport through ac-driven nanoscale systems. A main finding is that in molecular wires and coherently coupled quantum dots, the influence of an ac field can be used to selectively suppress both the current and its noise. Investigating the *relative* noise level characterized by the Fano factor has revealed that the current suppression is accompanied by a noise maximum and two remarkably low minima. These phenomena can be used to devise novel current sources with *a priori* controllable noise levels.

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