Asymmetric Quantum Shot Noise in Quantum Dots

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We analyze the frequency-dependent noise of a current through a quantum dot which is coupled to Fermi leads and which is in the Coulomb blockade regime. We show that the asymmetric shot noise as function of frequency shows steps and becomes super-Poissonian. This provides experimental access to the quantum fluctuations of the current. We present an exact calculation for a single dot level and a perturbative evaluation of the noise in Born approximation (sequential tunneling regime but without Markov approximation) for the general case of many levels with charging interaction.

The shot noise is a striking consequence of charge quantization and allows to characterize the transport of individual electrons \([1]\). The symmetry of the noise \(S(\omega)\) is important: For a classical stationary system, the noise (for autocorrelations) is always symmetric in the frequency \(\omega\). However, for a quantum system, the noise can by asymmetric due to the non-commutativity of current operators at different times. It was recently found that such an asymmetric noise can be detected since the noise frequency \(\omega\) corresponds to a quantum of energy \(\hbar \omega\) which is transferred from the system to the measurement apparatus \([2, 3, 4]\) which was demonstrated experimentally \([5]\). This means that antisymmetric quantum effects in the noise can be measured and isolated from the classical (symmetric) effects. We show that striking antisymmetric effects appear in the shot noise \(S(\omega)\) of a quantum dot with steps as function of \(\omega\), giving a super-Poissonian Fano factor. Our analysis is based on a perturbative approach which remains valid in the quantum limit with large \(\omega\) (where a Markov approximation typically invoked would not be valid). We confirm our perturbative results by an exact calculation of the noise for a dot with a single level. We note that quantum dots are good candidates for an experimental test of our predictions since such systems have been studied extensively over the years, both experimentally and theoretically \([4, 5, 6, 7, 8, 9, 10, 11]\).

We consider the operator \(I_{l'}\) which describes the current in a lead \(l\). We define the current noise,

\[
S_{\text{ll}}(\omega) = \int_{-\infty}^{\infty} dt \, e^{i \omega t} \left[ I_{l}(t) I_{l'}(t) - \langle I_{l}(t) \rangle \langle I_{l'}(t) \rangle \right],
\]

to illustrate the presence of asymmetric shot noise contributions due to quantum effects, we consider now a concrete system of a quantum dot in the Coulomb blockade regime \([12]\), coupled to Fermi leads \(l = 1, 2, ...\) at chemical potentials \(\mu_l\). When only a single dot level is present, the noise can be calculated even exactly \([6]\) (see below). This is however not possible for systems with many levels and charging interaction, for which we now develop a perturbative approach. We assume weak coupling such that current and noise are dominated by the sequential tunneling (ST) contributions, valid for \(kT > \gamma\) with temperature \(T\) and level width \(\gamma\). We model the combined system with the Hamiltonian

\[
H = H_{\text{lead}} + H_{\text{d}} + H_{\text{T}},
\]

where \(H_{\text{d}}\) describes leads, dot, and the tunnel coupling between leads and dot, resp., and with \(H_{\text{T}} = H_{\text{lead}} + H_{\text{d}}\). We let

\[
H_{\text{T}} = \sum_{lk\sigma} \epsilon_{lk} c_{lk\sigma}^\dagger c_{lk\sigma},
\]

where \(c_{lk\sigma}\) creates an electron in lead \(l\) with orbital state \(k\), spin \(\sigma\), and energy \(\epsilon_{lk}\). The electronic dot states \(|n\rangle\) are described by \(H_{\text{d}}|n\rangle = E_n |n\rangle\), including charging and interaction energies. We use the standard tunneling Hamiltonian

\[
H_{\text{T}} = \sum_{ljk\sigma} t_{ljk}^\dagger c_{lk\sigma}^\dagger d_{j\sigma}^\dagger + \text{H.c.},
\]

where \(d_{j\sigma}\) creates an electron on the dot with orbital state \(j\) and spin \(\sigma\). The state of the combined system is given by the full density matrix \(\rho\), while the electronic states of the dot are described by the reduced density matrix, \(\rho_{\text{SS}} = Tr_{\text{R}} \rho\), with the trace taken over the leads. We assume that at some initial time \(t_0\) the full density matrix factorizes, \(\rho(t_0) = \rho_{\text{SS}}^0 \rho_{\text{R}}^0\) with the equilibrium density matrix of the leads, \(\rho_{\text{R}}^0\). From the von Neumann equation \(\dot{\rho} = -i[H, \rho]\) one finds

\[
\dot{\rho}_{\text{SS}}(t) = -i L_{\text{SS}} \rho_{\text{SS}}(t) - \int_{t_0}^{t} dt' M(t') \rho_{\text{SS}}(t-t'),
\]

where the kernel \(M\) is the self-energy superoperator. Since we consider the weak coupling regime, we proceed with a systematic lowest-order expansion in \(H_{\text{T}}\). We obtain \(M(\tau) = Tr_{\text{R}} L_{\text{T}} e^{-i \tau \omega} L_{\text{T}} \rho_{\text{R}}^0\), where we define
We obtain the noise correlation in the ST regime, \( t - t_0 = 1 \) we get \( Q_p \) and the current.

\[
M(\omega) = -i L_S - M(\omega) \text{ with the lower boundary of the Laplace transformation shifted to } t_0. \text{ We take } t_0 \to -\infty \text{ and assume that the system has relaxed at the much later time } t = 0 \text{ into its stationary state } \tilde{\rho}_S = \rho_S(0) = \lim_{\omega \to 0} (-i \omega) \rho_S(\omega). \text{ We multiply Eq. (1)} \text{ by } -i \omega, \text{ take } \omega \to 0, \text{ and find the equation } M(\omega) \tilde{\rho}_S = 0 \text{ from which we get } \tilde{\rho}_S.
\]

Current. We calculate the current \( I_t \) flowing from the dot into lead \( l \) and vice versa. The current operators are \( I_t(t) = (-1)^l cQ_l(t) = (-1)^l i e[H_T, q(t)] \) where \( q_l = \sum_{\kappa} c_{\kappa l} c_{\kappa l} \) is the number of electrons in lead \( l \). We choose the sign of \( I_t \) such that \( \langle I_t \rangle = \langle I_\bar{t} \rangle \) in the case of two leads. We now introduce the projectors \( P = \rho_{lR}^0 T_R \) and \( Q = \mathbb{1} - P \) with the properties \( P L_T = P = P L_T \) and \( P L_0 = L_0 P \). We evaluate \( I_t \) by inserting \( P + Q = \mathbb{1} \) and find \( \langle I_t(t) \rangle = Tr[I_t Q e^{-i L_0 T_L(t-t_0)}] \tilde{\rho}(t_0) = -i Tr[I_t l^0 \Delta_{L_T(t-t_0)} T_L P] \tilde{\rho}(t_0) = -i Tr[I_t e^{-i L_0 T_L} T_L P] \tilde{\rho}(t_0). \) Note that these superoperators act only on the dot space, which considerably simplifies further evaluations.

In the ST regime, the current is

\[
\langle I_t \rangle = Tr[I_l W_l^t(\omega) = 0] \tilde{\rho}_S.
\]

This implies that the superoperator \( W_l^t \) corresponds to the current.

Quantum shot noise. We evaluate the noise \[ Eq. (11) \] in lowest order in \( H_T \), but without any further approximation. It is sufficient to consider \( t > 0 \), since \( \langle I_t(t) I_{\bar{t}} \rangle = \langle I_t(t) I_{\bar{t}} \rangle^* \). Using again \( P + Q = \mathbb{1} \) we get \( \langle I_t(t) I_{\bar{t}} \rangle = Tr[I_t Q e^{-i L_0 T_L} T_L P] \tilde{\rho}(t_0) \) and \( W_l^c(t) = Tr[I_t e^{-i L_0 T_L} T_L P] \tilde{\rho}(t_0). \) Note that these superoperators act only on the dot space, which considerably simplifies further evaluations.

We now return to the exact expression of the noise in Born approximation \[ Eq. (11) \] and explicitly calculate the matrix elements of the various superoperators.

\[
-M(t) e^{i L_0 t} \rho_S = \sum_l (G_{l+}^l \rho_S - g_{l+}^l \rho_S) + h.c.,
\]

\[
W_l^c(t) e^{i L_0 t} = (-1)^l e^{i(\omega t + g_l^t)}.
\]

With the principal values \[ Eq. (12) \] of the dot spectrum; one only needs to evaluate simple algebraic expressions.

\[
G_{l+,l}^c(\omega) \cong G_{l+,l}^c(\Delta_{l0} - \epsilon) + \frac{\epsilon}{\pi},
\]

\[
G_{l+,l}^c(\omega) \cong \mu^l_{\text{max}} \left( \frac{1}{\pi} \right),
\]

where \( \Delta_{nb} = E_b - E_n \), and \( \mu^l_{\text{max}} = \text{log}(2\pi k T/|[1 \mp 1] \epsilon_c/2 + \Delta_{nb} - \omega|) + \text{Re} \psi \left[ \frac{1}{2} + i \Delta_{nb} + \omega - \mu/2 \right] \). Here, \( \psi \) is the digamma function. The terms \( \mu^+ - \mu^- \) arise from the principal values \[ Eq. (15) \] of the dot spectrum; one only needs to evaluate simple algebraic expressions.

We now identify the regime where the asymmetric noise properties become most apparent. Asymmetries arise from the \( |\omega| > kT \), with steps occurring at \( |\omega| \approx |\Delta_{nb} - \mu| \) (see below). In this regime, the Markov approximation breaks down \[ Eq. (10) \] and the noise probes
consider the coherent non-perturbative regime of strong coupling to the leads in the quantum limit of large frequencies, \( \omega > \gamma > kT \). We obtain the shot noise

\[ S_n^\downarrow(\omega) = 2 \sum_{\nu, \pm} \frac{\gamma_\nu \gamma_\nu'}{2\gamma} \theta(\omega \pm \mu_\nu \mp \mu_\nu) |h(\mu_\nu') - h(\mu_\nu \mp \omega)|, \]

where \( h(\epsilon) = \arctan[(\epsilon - E_\uparrow)/\gamma] \). Note that the noise shows steps at \( \omega = \pm |E_\uparrow - \mu_\nu| \) with width \( \gamma \). Furthermore, for \( \omega > |E_\uparrow - \mu_\nu|, \Delta \mu \), the noise is asymmetric and saturates at \( S_n^\downarrow(\omega) = e^2\gamma_\nu \), while \( S_n^\uparrow(\omega) = 0 \).

Let us now consider the ST regime \( kT > \gamma \) in the exact solution. For \( \omega > \gamma \), we then find

\[ S_n^\downarrow(\omega) = 2 \sum_{\nu, \pm} \frac{\gamma_\nu \gamma_\nu'}{2\gamma} \left[ \delta_{1, \mp 1} \pm f_\nu(E_\uparrow) \right] \left[ \delta_{1, \mp 1} \mp f_\nu(E_\downarrow \mp \omega) \right]. \]

Again, the noise shows a pronounced asymmetry. We can now compare Eq. (11) with the noise obtained in the perturbative approximation [Eq. (1)] and find that they agree. We further consider \( \omega > \Delta \mu + kT \) in Eq. (11) such that \( f_\nu(E_\uparrow + \omega) = 0 \) and \( f_\nu(E_\downarrow - \omega) = 1 \), leaving \( f_\nu(E_\uparrow) \) unrestricted. In this case, the (asymmetric) shot noise is

\[ S_n^\downarrow(\omega) = e^2\gamma_\nu, \]

whereas \( S_n^\uparrow(\omega) = S_n^\downarrow(\pm \omega) = 0 \); this is the same result as we have found for strong coupling [Eq. (11)]. The interpretation is that for \( S_n^\downarrow(\omega) \), the detector absorbs energy \( \omega \), which, however, cannot be provided by any tunneling process. On the other hand, for \( S_n^\uparrow(\omega) \) the detector provides energy \( \omega \). Thus, if the dot is empty, an electron with energy \( E_\uparrow - \omega \) can tunnel from the Fermi sea into the dot, and, if the dot is filled, an electron can tunnel from the dot into an unoccupied lead state of energy \( E_\downarrow + \omega \), see Fig. 1(a). In both cases, the tunneling occurs with rate \( \gamma_\nu \) and thus the contribution to the autocorrelation is \( e^2\gamma_\nu \delta(t) \). Note that for \( |E_\uparrow - \mu_\nu| > kT \), the noise is \( S_n^\downarrow(\omega) = e^2(\gamma_\nu \gamma_\nu' + \gamma_\nu' \gamma_\nu)/2 \). Thus, for large \( \omega \), the frequency dependent Fano factor, \( F_{11}(\omega) = S_{11}(\omega)/e(I) \), is 2 for \( \gamma_\nu = \gamma_\nu' \), and can even become larger for \( \gamma_\nu > \gamma_\nu' \), in contrast to the Markovian case where we find it to be 1. Thus, we find that the quantum shot noise is super-Poissonian.

Moreover, away from the ST regime, say for \( E_\uparrow + kT > \mu_\nu \), the dot remains in state \( |0\rangle \) and only a small (higher-order in \( H_T \)) cotunneling current \( I \) flows through the dot. However, the noise can still be of lower order, it is \( S_n^\downarrow(\omega) = e^2\gamma_\nu f_\nu(E_\uparrow - \omega) \) for large \( |\omega| \), resulting in a large Fano factor \( F_{11}(\omega) \) and super-Poissonian shot noise.

**Dot with two or more levels.** Second, we consider the regime where the state \( |\downarrow\rangle \) becomes relevant and charging interaction enters (here no exact solution is available). We consider a small Zeeman splitting such that \( \mu_\uparrow > E_\uparrow > \mu_\downarrow \) and \( f_\nu(E_\uparrow) \approx f_\nu(E_\downarrow) \), see Fig. 1(b). Using Eq. (11), we can calculate the noise \( S_n^\downarrow(\omega) \) and plot it in Fig. 2 (solid line). For large \( |\omega| \), such that \( f_\nu(E_\uparrow + |\omega|) = 0 \) and \( f_\nu(E_\downarrow - |\omega|) = 1 \), the noise vanishes for \( \omega < 0 \) while for
D. Loss, E.V. Sukhorukov, Phys. Rev. Lett. and within Markov approximation, S. (Asymmetric noise at frequencies up to 90 GHz has been shot noise regime \( \Delta \)). More generally, for the weaker assumption \( \omega > 0 \) it saturates at

\[
S_{ll}^{\uparrow}(\omega) = 2e^2 \gamma_1 \frac{\gamma_1 + \gamma_2}{\gamma_1 [1 + f_{l}(E_{\uparrow})] + \gamma_2 [1 + f_{l}(E_{\uparrow})]}.
\] (13)

More generally, for the weaker assumption \( |\omega| > \gamma_1 \), the numerator in Eq. (13) becomes

\[
\frac{1}{2} \sum_{l',\pm,\sigma} \gamma_{ll'} [\delta_{l',\pm1} \pm f_{l'}(E_{\sigma})] [\delta_{l',\pm1} \pm f_{l'}(E_{\sigma} \pm \omega)].
\]

Thus, the noise \( S_{ll}^{\uparrow} \) shows four steps at \( \omega = \pm(\mu_1 - E_{\uparrow}) \), corresponding to the dotted lines in Fig. 1(b). The interpretation is that for increasing \( \omega \), more energy is available and more tunneling processes are allowed. Namely, if \( \omega \geq -(\mu_1 - E_{\uparrow}) \), an electron with spin \( \uparrow \) from lead 1 can emit energy \( \omega \) and tunnel onto the dot, and if \( \omega \geq \mu_1 - E_{\uparrow} \), an electron on the dot with spin \( \uparrow \) can absorb energy \( \omega \) and tunnel into lead 1. Analogous processes occur for spin \( \downarrow \). The steps in \( S_{ll}^{\uparrow} \) are broadened due to \( \omega \), and the step \( \Delta \) is \( \propto \tanh[(\omega - \omega_i)/2kT] \). For \( \Delta \mu > kT, \Delta \), i.e., \( f_{1}(E_{\sigma}) = \delta_{11} \), the height of the steps in the Fano factor \( F_{11}(\omega) \) is \( \frac{1}{2} \) for \( \omega < 0 \) and \( \gamma_1/2\gamma_2 \) for \( \omega > 0 \). (These heights change for spin-dependent tunneling, \( \gamma_{\uparrow} \neq \gamma_{\downarrow} \).) Next, we consider an intermediate Zeeman splitting, \( E_{l} > \mu_1 + kT \). In this regime (c), the dot is either in state \(|0\rangle \) or \(|\uparrow\rangle \), while the state \(|\downarrow\rangle \) is never occupied and so no additional tunneling process occurs for \( \omega \geq -(E_{l} - \mu_1) \). Thus, the steps in the noise are at \( \omega = \pm(\mu_1 - E_{\uparrow}) \) and at \( \omega = E_{l} - \mu_1 \), see Fig. 2(dashed line). For large \( \omega \), the noise saturates at

\[
S_{ll}^{\uparrow}(\omega) = e^2 \sum_{l=1,2} \gamma_{ll'}^{\uparrow} \frac{\gamma_{\uparrow} + \gamma_{\downarrow}[1 - f_{l'}(E_{\uparrow})]}{\gamma_{\uparrow} + \gamma_{\downarrow}}. \tag{14}
\]

Here, we have allowed for spin-dependent tunneling. If we exclude the contributions involving state \( |\downarrow\rangle \) by setting \( \gamma_{\downarrow} = 0 \), we recover Eq. 12. Generally, we see that the shot noise \( S_{ll}(\omega) \) of a quantum dot consists of a series of steps and is monotonically increasing, apart from features near \( \omega = 0 \). Each dot level with energy (chemical potential of the dot) \( E_{j} \) gives rise to steps at \( \pm(\mu_1 - E_{j}) \) if the level is inside the bias window, \( \mu_1 + kT > E_{j} > \mu_2 - kT \), and to a single step at \( |\mu_j - E_{j}| \) otherwise. We stress that such a highly asymmetric \( S_{ll}(\omega) \) can be observed with an appropriate measurement apparatus [2, 3, 4]. For sufficiently large \( |\omega| \), the antisymmetric contribution becomes \( \frac{1}{2} [S_{ll}(\omega) - S_{ll}(-\omega)] = 1 2 3 4 |\omega| ) S_{ll}(\omega) ) |(\omega)| and is, e.g., given by Eqs. 13 - 14.

In conclusion, we have derived the asymmetric shot noise of a quantum dot exactly for a single dot level and in the weak coupling regime for many levels. We have shown that the shot noise exhibits strong asymmetric and super-Poissonian effects in the quantum limit. We thank F. Marquardt, W. Belzig, G. Burkard, A. Cottet, J.C. Egues, H. Gassman, D. Saraga, and C. Schönenberger for discussions. We acknowledge support from the Swiss NSF, NCCR Nanoscience Basel, DARPA, and ARO.

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[15] The formal solution of Eq. 2 corresponds to resumming an infinite number of terms. Since we evaluate the self-energy \( \mathcal{M} \) in leading order, we only resum a certain subset of the higher order terms in \( \rho_{\uparrow}(\omega) \).

Figure 2: The Fano factor \( F_{11}(\omega) = S_{11}(\omega)/e(I) \) in the shot noise regime \( \Delta \mu > kT \) as function of noise frequency \( \omega \). (Asymmetric noise at frequencies up to 90 GHz has been measured [2].) We consider \( T = 100 \text{mK}, \Delta \mu/e = 460 \text{µV}, E_{l} = (\mu_1 + \mu_2)/2 \), \( \gamma_1 = \gamma_2 = 5 \times 10^{9} \text{s}^{-1} \), and \( g = 2 \). We use the full expression for the noise \( S_{11}^{\text{Markov}} \) (Eq. 4) (solid line) and within Markov approximation, \( S_{11}^{\text{Markov}} \) (dotted line), for \( B = 1 \text{T} \) [see Fig. 1(b)], thus \( \Delta_l = \Delta \mu/4 \) and \( (I) = 530 \text{pA} \). We also show \( S_{ll}^{\uparrow} \) (dashed line), being strongly asymmetric, where \( B = 3 \text{T}, \Delta_l = 3 \Delta \mu/4 \), and \( (I) = 400 \text{pA} \). The dip near \( \omega = 0 \) is due to the charging effect of the dot, while the steps at \( \omega_1 \) (see text) arise from additional transitions for increasing \( \omega \) and provide a striking effect in the quantum shot noise. Note that these steps disappear when the noise \( S_{ll}^{\uparrow} \) is symmetrized (some features remain for \( \gamma_1 \neq \gamma_2 \)).
[16] Evaluating $M(\tau)$ explicitly, we find that it decays on a time scale $\tau_c \sim 1/kT$, i.e., the correlations induced in the leads decay within $\tau_c$. Thus, the Markov approximation is justified for $\omega \tau_c, \gamma \tau_c \ll 1$.

[17] In Born-Markov approximation, the autocorrelation function is always symmetric in $\omega$ for operators $X$ with $[X, H_0] = [X, \hat{\rho}_R] = 0$.

[18] This linear map $G \mapsto g$ is the identity on operators, however, here $G$ is a superoperator. With this notation, Eq. (5) is reminiscent of the Lindblad form, $\sum_i [A_i, \rho S - A_i^\dagger A_i \rho S] + h.c.$

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