Asymmetric Quantum Shot Noise in Quantum Dots

Hans-Andreas Engel and Daniel Loss

Department of Physics and Astronomy, University of Basel, Klingelbergstrasse 82, CH-4056 Basel, Switzerland

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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 a function of detection frequency, shows steps and becomes super-Poissonian. This provides experimental access to the quantum fluctuations of the current. We present an exact calculation of the noise 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.

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Shot noise is a striking consequence of charge quantization and characterizes the transport of individual electrons [1]. The theoretical description of noise is an active field of research and so far has focused on symmetrized noise, $S^{\rm sym}(\omega)$. However, it was recently found that asymmetric noise, $S(\omega)$, can also be detected since the noise frequency ω corresponds to a quantum of energy $\hbar\omega$ being transferred from the measurement apparatus to the system [2–7], which has been demonstrated experimentally [8,9]. In this Letter we analyze the asymmetric shot noise occurring in nonequilibrium quantum systems [10]. We calculate the asymmetric noise of a quantum dot for the first time and find striking asymmetric effects, namely, steps in $S(\omega)$. Further, the Fano factor (ratio of noise to current) becomes super-Poissonian for large frequencies. For an experimental test of these predictions, quantum dots are good candidates since they have been studied extensively over the years, both experimentally and theoretically [11–16]. Our analysis of noise is based on a systematic perturbative approach within a standard superoperator formalism. We go beyond previous calculations for dots which were done for small detection frequencies ω with respect to bias and temperature (allowing for a classical description of noise), and our results remain valid in the quantum limit of large ω. (In this limit, a Markov approximation typically invoked would not be valid, i.e., we take non-Markovian effects into account.) We obtain the asymmetric noise of a quantum dot in the sequential tunneling regime, including charging effects and allowing for an arbitrary level spectrum. Further, we exactly calculate the dynamics of a dot with a single level and its asymmetric noise. This confirms our perturbative derivation and supports our general results.

Asymmetric noise.— We consider the operator I_l which describes the current in some lead l. We define the current noise,

$$S_{ll'}(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t}[\langle I_l(t)I_{l'}\rangle - \langle I_l\rangle\langle I_{l'}\rangle], \qquad (1)$$

in terms of the (nonsymmetrized) correlation function, $\langle I_l(t)I_{l'}\rangle = {\rm Tr} I_l(t)I_{l'}\bar{\rho}$. Here, $\bar{\rho}$ is the stationary density matrix (of the full quantum system). Note that $I_l(t)I_{l'}$ is not Hermitian and thus does not correspond to an observable. One can avoid this non-Hermitian operator by arguing heuristically that Eq. (1) is "unphysical" and by considering the correlation function in terms of the symmetrized operator $\frac{1}{2}[I_l(t)I_{l'} + I_{l'}I_l(t)]$ instead [17], leading to symmetrize noise $S^{\rm sym}_{ll'}(\omega)$. However, we have $S_{ll'}(\omega) = S_{l'l}(\omega)^*$, since I_l is Hermitian and $\bar{\rho}$ stationary, so $S_{ll}(\omega)$ is a real quantity. Thus, $S_{ll}(\omega)$ can be regarded as an observable. This interpretation is justified by setups which can measure $S_{ll}(\omega)$ [2–9].

Quantum dots.— To illustrate the presence of asymmetric shot noise contributions due to quantum effects, we consider a concrete system of a quantum dot in the Coulomb blockade regime [18] coupled to Fermi leads $l = 1, 2, \ldots$ at chemical potentials μ_l. When only a single dot level is present, the noise can be calculated exactly [12] (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 γ. We model the combined system with the Hamiltonian $H = H_{\rm lead} + H_{\rm d} + H_T$, which describes leads, dot, and the tunnel coupling between leads and dot, respectively, and with $H_0 = H_{\rm lead} + H_{\rm d}$. We let $H_{\rm lead} = \sum_{lk\sigma}\epsilon_{lk}c^\dagger_{lk\sigma}c_{lk\sigma}$, where $c^\dagger_{lk\sigma}$ creates an electron in lead l with orbital state k, spin σ, and energy ϵ_{lk}. The electronic dot states $|n\rangle$ are described by $H_{\rm d}|n\rangle = E_n|n\rangle$, including charging and interaction energies [18,19]. We use the standard tunneling Hamiltonian $H_T = \sum_{lpk\sigma} t^\sigma_{lp} c^\dagger_{lk\sigma} d_{p\sigma} + {\rm H.c.}$, with tunneling amplitude t^σ_{lp} and where $d^\dagger_{p\sigma}$ creates an electron on the dot with orbital state p and spin σ. The state of the combined system is given by the full density matrix ρ, while the electronic states of the dot are described by the reduced density matrix, $\rho_{\rm d} = {\rm Tr}_{\rm R}\rho$, where the trace is taken over the leads. We assume that at some initial time t_0 the full density matrix factorizes, $\rho(t_0) = \rho^0_{\rm d}\rho^0_{\rm R}$, with the equilibrium density matrix of the leads, $\rho^0_{\rm R}$. From the

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von Neumann equation $\dot{\rho} = -i[H, \rho]$, one finds [20] the generalized master equation for the reduced density matrix, $\dot{\rho}_{\rm d}(t) = -iL_{\rm d}\rho_{\rm d}(t) - \int_{t_0}^{t} dt'\hat{M}(t')\rho_{\rm d}(t-t')$. Here, the kernel $\hat{M}$ is the self-energy superoperator, and we define the superoperators $L_iX = [H_i, X]$. Since we consider the weak coupling regime, we proceed with a lowest-order expansion in H_T and obtain $\hat{M}(t) = {\rm Tr}_{\rm R} L_T e^{-iL_0 t} L_T \rho_{\rm R}^0$. In the following, we work in Laplace space, $f(t) \mapsto f(\omega) = \int_0^\infty dt e^{i\omega t} f(t)$ (we take ${\rm Im}\omega > 0$ but our results remain well defined for ${\rm Im}\omega \to 0$). Then, the time evolution of $\rho_{\rm d}$ reads

$$-\rho_{\rm d}(t_0) - i\omega\rho_{\rm d}(\omega) = \mathcal{M}(\omega)\rho_{\rm d}(\omega), \tag{2}$$

with $\mathcal{M}(\omega) = -iL_{\rm d} - \hat{M}(\omega)$ and with the lower boundary of the Laplace transform shifted to t_0. We take $t_0 \to -\infty$ and assume that the system has relaxed at the much later time $t = 0$ into its stationary state $\bar{\rho}_{\rm d} = \rho_{\rm d}(0) = \lim_{\omega\to 0}(-i\omega)\rho_{\rm d}(\omega)$. Using Eq. (2), we find $\mathcal{M}(0)\bar{\rho}_{\rm d} = 0$, from which we get $\bar{\rho}_{\rm d}$.

Current.— We calculate the current I_l flowing from the dot into lead l and vice versa. The current operator is $I_l(t) = (-1)^l e\dot{q}_l(t)$ where q_l is the number of electrons in lead l. We choose the sign of I_l such that $\langle I_1\rangle = \langle I_2\rangle$ in the case of two leads. We evaluate $\langle I_l\rangle$ in lowest order of H_T [21] and introduce $W_l^{\rm f} = W_l^> + W_l^<$ with $W_l^>(t) = -i{\rm Tr}_{\rm R} I_l e^{-iL_0 t} L_T \rho_{\rm R}^0$ and $W_l^<(t) = -i{\rm Tr}_{\rm R} L_T e^{-iL_0 t} I_l \rho_{\rm R}^0$, and $W_{l,l'}(t) = {\rm Tr}_{\rm R} I_l e^{-iL_0 t} I_{l'} \rho_{\rm R}^0$. These superoperators act only on the dot space. In the ST regime, we find

$$\langle I_l\rangle = {\rm Tr}_{\rm d} W_l^{\rm f}(\omega = 0)\bar{\rho}_{\rm d}. \tag{3}$$

This indicates that the superoperator $W_l^{\rm f}$ accounts for the current through the dot.

Quantum shot noise.— We now evaluate the noise [Eq. (1)] to lowest order in H_T but without any further approximation. Using some standard identities [20,21], we factor out the conditional time evolution $\rho^{\rm c}(t) := {\rm Tr}_{\rm R} e^{-iLt}\rho_{\rm R}^0$, which propagates an arbitrary initial dot state by time t. We see that $\rho^{\rm c}$ is the formal solution of the master equation [Eq. (2)] with initial value $\mathbb{1}$, thus $\rho^{\rm c}(\omega) = -[i\omega + \mathcal{M}(\omega)]^{-1}$ [21]. We obtain the noise correlation in the ST regime [21,22],

$$\begin{aligned} S_{ll'}(\omega) = 2{\rm Tr}_{\rm d}\Big\{ & W_l^{\rm f}(\omega)\rho_{\rm d}^{\rm c}(\omega)[W_{l'}^>(0) + W_{l'}^<(\omega)] \\ & + W_{l'}^{\rm f}(-\omega)\rho_{\rm d}^{\rm c}(-\omega)[\tilde{W}_l^>(0) + \tilde{W}_l^<(-\omega)] \\ & + W_{l,l'}(\omega) + \tilde{W}_{l',l}(-\omega)\Big\}\bar{\rho}_{\rm d}. \end{aligned} \tag{4}$$

Here, ω is real and the limit $\omega \to 0$ is well behaved. For a superoperator $S(t)$, we have defined $\tilde{S}(t)$ such that $[S(t)A]^\dagger = \tilde{S}(t)A^\dagger$. In deriving Eqs. (3) and (4), we have made no Markov approximation where we would evaluate $\hat{M}(t)e^{itL_0}$ at $\omega = 0$ and equivalently for the other superoperators [21].

We now return to the exact expression for the noise in Born approximation [Eq. (4)] and explicitly calculate the matrix elements of the various superoperators,

$$-\hat{M}(t)e^{itL_0}\rho_{\rm d} = \sum_l (G_+^l\rho_{\rm d} - g_+^l\rho_{\rm d}) + {\rm H.c.}, \tag{5}$$

$$W_l^{<(>)}(t)e^{itL_0} = (-1)^l e\Big(\overset{(\sim)}{G_-^l} \mp g_-^l\Big), \tag{6}$$

$$W_{l,l'}(t)e^{itL_0} = \delta_{ll'}e^2 g_+^l, \tag{7}$$

with $G_\pm^l = G^{l,{\rm out}}(t) \pm G^{l,{\rm in}}(t)$ and $g_\pm^l = \sum_{bn}|b\rangle \times \langle n|{\rm Tr}_{\rm d}\{G_\pm^l|n\rangle\langle b|\}$ [23]. We define $t_{nm}^{l\sigma} = \sqrt{2\pi\nu_{l\sigma}}\sum_p t_{lp}^\sigma\langle n|d_{p\sigma}|m\rangle$, with spin-dependent density of states $\nu_{l\sigma}$ in lead l. The matrix elements, $S_{bc|nm} := \langle b|(S|n\rangle\langle m|)|c\rangle$, of the remaining superoperators are

$$G_{bc|nm}^{l,{\rm in}}(\omega) = \sum_\sigma \frac{t_{mc}^{l\sigma}t_{nb}^{l\sigma*}}{2}\Big[f_l(\Delta_{bn} - \omega) + \frac{ip_{bn}^+}{\pi}\Big], \tag{8}$$

$$G_{bc|nm}^{l,{\rm out}}(\omega) = \sum_\sigma \frac{t_{mc}^{l\sigma*}t_{bn}^{l\sigma}}{2}\Big[1 - f_l(\Delta_{nb} + \omega) + \frac{ip_{nb}^-}{\pi}\Big], \tag{9}$$

with $\Delta_{bn} = E_b - E_n$, which contains the charging energy, and $p_{bn}^\pm = \log\{2\pi kT/[(1 \mp 1)\epsilon_c/2 \pm \Delta_{bn} - \omega]\} + {\rm Re}\psi[\frac{1}{2} + i(\Delta_{bn} \mp \omega - \mu_l)/2\pi kT]$ and with digamma function ψ and bandwidth cutoff ϵ_c. If we neglect ω with respect to the large energies Δ_{bn} and $\epsilon_c - \Delta_{nb}$, the first term of $p^\pm$ (and thus ϵ_c) drops out in $S_{ll'}(\omega)$. We note that the contribution corresponding to Eq. (7) has been calculated for the symmetrized noise of a single electron transistor with a continuous spectrum, using a phenomenological Langevin approach [24]. With our results, Eqs. (4) and (5)–(9), it is straightforward to find $S_{ll'}(\omega)$ for an arbitrary 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 ω dependence of Eqs. (8) and (9), i.e., are most prominent for $|\omega| > kT$, with steps occurring at $|\omega| \simeq |\Delta_{bn} - \mu_l|$ (see below). In this regime, the Markov approximation breaks down (it changes the noise, which for $l = l'$ becomes symmetrized, $S_{ll}^{\rm Mkov}(\omega) = S_{ll}^{\rm Mkov}(-\omega)$) and noise probes non-Markovian effects. For the noise [Eq. (4)], only the two last terms are relevant, since they are of order γ while the other terms are of order γ^2/ω and can be neglected. This is because multiple tunneling processes [described by $\rho^{\rm c}(\omega)$] do not occur on the short time scales corresponding to large ω. Thus, only the individual (uncorrelated) tunneling events contribute, leading to shot noise.

Next, we discuss specific cases, see Fig. 1, where a dot is coupled to two leads $l = 1, 2$ and a voltage bias $\Delta\mu = \mu_1 - \mu_2$ is applied. We assume single energy level spacing and Coulomb charging energy larger than temperature, bias, and noise frequency. We consider the dot state $|0\rangle$ with an even number of electrons and with $E_0 = 0$, and the states $|\sigma\rangle$ where an electron with spin $\sigma = \uparrow, \downarrow$ is added to the dot [21]. For an applied magnetic field B, the Zeeman splitting is $\Delta_z = g\mu_B B = E_\downarrow - E_\uparrow > 0$. We consider the ST regime, $\mu_1 > E_\uparrow > \mu_2$, and define the tunneling rates $\gamma_l^\sigma = |t_{0\sigma}^{l\sigma}|^2$.

Dot with single level.— First, we assume a large Zeeman splitting such that only the spin ground state $|\uparrow\rangle$ is relevant, see Fig. 1(a), and we can omit the index $\uparrow$. Since in this regime only one dot level is involved, there are no charging effects between different levels. Thus, $H_d = E_\uparrow d^\dagger d$ and so the full Hamiltonian H is bilinear and can be solved exactly. The symmetrized noise was calculated for this system and discussed for $\omega = 0$ [12]. We now calculate the asymmetric noise [Eq. (1)] for finite ω exactly. For this, we solve the Heisenberg equations for $d(t)$ and $c_{lk}(t)$ and find the current operator, $I_l(t)/e(-1)^l = \sum_{kl'k'}[j_{k'} c^\dagger_{l'k'} c_{lk} + \text{H.c.}] + \sum_{l'k'l''k''}(\gamma_l/|t_l|^2) j_{k'} j^*_{k''} c^\dagger_{l'k'} c_{l''k''}$. Here, the lead operators c_{lk} are evaluated at time t_0 (i.e., when the leads are isolated and at equilibrium) and we have defined $j_{k'} = i t_l^* t_{l'} e^{i(\epsilon_{l'k'} - \epsilon_{lk})(t-t_0)}/(\epsilon_{l'k'} - E_\uparrow - i\gamma)$ and $\gamma = (\gamma_1 + \gamma_2)/2$. Now we insert $I_l(t)$ into Eq. (1) and readily obtain the asymmetric noise, containing all quantum effects. We consider the coherent nonperturbative regime of strong coupling to the leads in the quantum limit of large frequencies, $\omega > \gamma > kT$. We obtain the shot noise

$$S^{a}_{ll}(\omega) = \sum_{l',\pm} \frac{\pm e^2 \gamma_1 \gamma_{l'}}{2\pi\gamma} \theta(\omega \pm \mu_{l'} \mp \mu_l)[h(\mu_{l'}) - h(\mu_l \mp \omega)], \tag{10}$$

where $h(\epsilon) = \arctan[(\epsilon - E_\uparrow)/\gamma]$. Note that the noise shows steps at $\omega = \pm|E_\uparrow - \mu_l|$ with width γ. Furthermore, for $\omega > |E_\uparrow - \mu_l|, \Delta\mu$, the noise is asymmetric and saturates at $S^a_{ll}(\omega) = e^2\gamma_l$, while $S^a_{ll}(-\omega) = 0$.

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

$$S^{a}_{ll}(\omega) = \sum_{l',\pm} \frac{e^2 \gamma_l \gamma_{l'}}{2\gamma} [\delta_{1,\mp 1} \pm f_{l'}(E_\uparrow)][\delta_{1,\pm 1} \mp f_l(E_\uparrow \pm \omega)]. \tag{11}$$

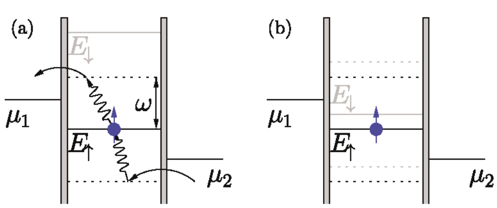

FIG. 1 (color online). Quantum dot coupled to two leads and in the sequential tunneling regime. (a) Large Zeeman splitting, $\Delta_z > \Delta\mu + \omega$. When the dot is empty and $\omega \geq E_\uparrow - \mu_2$, an electron from lead 2 absorbs energy ω and tunnels for a short time onto the dot, contributing to the noise S_{22}. Similarly, for $\omega > \mu_1 - E_\uparrow$, the electron on the dot can tunnel into lead 1, contributing to S_{11}. (b) Smaller Zeeman splitting, here $\Delta_z = \Delta\mu/4$. When the energies $E_{\uparrow,\downarrow} \pm \omega$ (dotted lines) are aligned with $\mu_{1,2}$, the shot noise has a step; see Fig. 2 (solid line).

Again, the noise shows steps and a pronounced asymmetry. We can now compare Eq. (11) with the noise obtained in the perturbative approximation [Eq. (4)] and find that they agree. We further consider $\omega > \Delta\mu + kT$ in Eq. (11) such that $f_l(E_\uparrow + \omega) = 0$ and $f_l(E_\uparrow - \omega) = 1$, leaving $f_l(E_\uparrow)$ unrestricted. Then, the (asymmetric) shot noise is

$$S^a_{ll}(\omega) = e^2\gamma_l, \tag{12}$$

whereas $S^a_{ll}(-\omega) = S^a_{12}(\pm\omega) = 0$; this is the same result as we have found for strong coupling [Eq. (10)]. The interpretation is that for $S^a_{ll}(-\omega)$, the detector absorbs energy ω, which, however, cannot be provided by any tunneling process. On the other hand, for $S^a_{ll}(\omega)$ the detector provides energy ω. Thus, if the dot is empty, an electron with energy $E_\uparrow - \omega$ can tunnel from the Fermi sea l into the dot, and if the dot is filled, an electron can tunnel from the dot into an unoccupied lead state of energy $E_\uparrow + \omega$; see Fig. 1(a) [25]. Note that for $|E_\uparrow - \mu_l| > kT$, the noise is $S^a_{11}(\omega) = e\langle I\rangle(\gamma_1 + \gamma_2)/\gamma_2$. Thus, for large ω, the frequency-dependent Fano factor, $F_{11}(\omega) = S_{11}(\omega)/e\langle I\rangle$, is two for $\gamma_1 = \gamma_2$, and can even become larger for $\gamma_1 > \gamma_2$, in contrast to the Markovian case where we find it to be 1. Thus, we find that the quantum shot noise is *super-Poissonian* [26]. Moreover, away from the ST regime, say for $E_\uparrow + kT > \mu_l$, the dot always remains in state $|0\rangle$ and only a small (higher order in H_T) cotunneling current $\langle I\rangle$ flows through the dot [16]. However, the noise can still be of lower order; it is $S^a_{ll}(\omega) = e^2\gamma_l f_l(E_\uparrow - \omega)$ for large $|\omega|$, resulting in a large Fano factor $F_{ll}(\omega)$ 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 $\mu_1 > E_{\uparrow,\downarrow} > \mu_2$ and $f_l(E_\uparrow) \approx f_l(E_\downarrow)$; see Fig. 1(b). Using Eq. (4), we calculate the noise $S^b_{11}(\omega)$ and plot it in Fig. 2 (solid line). For large $|\omega|$, such that $f_l(E_\sigma + |\omega|) = 0$ and $f_l(E_\sigma - |\omega|) = 1$, the noise vanishes for $\omega < 0$ while for $\omega > 0$ it saturates at

$$S^b_{ll}(\omega) = 2e^2\gamma_l \frac{\gamma_1 + \gamma_2}{\gamma_1[1 + f_1(E_\uparrow)] + \gamma_2[1 + f_2(E_\uparrow)]}. \tag{13}$$

More generally, for the weaker assumption $|\omega| > \gamma$, the numerator in Eq. (13) becomes $\frac{1}{2}\sum_{l',\pm,\sigma}\gamma_{l'}[\delta_{1,\mp 1} \pm f_{l'}(E_\uparrow)][\delta_{1,\pm 1} \mp f_l(E_\sigma \pm \omega)]$. Thus, S^b_{11} shows four steps at $\omega_k = \pm(\mu_1 - E_{\uparrow,\downarrow})$, indicated by the dotted lines in Fig. 1(b), since for increasing ω, more energy is available and more tunneling processes are allowed. The steps in S^b_{ll} are broadened due to temperature, and the step is $\propto \tanh[(\omega - \omega_k)/2kT]$. Next, we consider an intermediate Zeeman splitting, $E_\downarrow > \mu_1 + kT$. In this regime (c), the dot is either in state $|0\rangle$ or $|\uparrow\rangle$, while the state $|\uparrow\rangle$ is never occupied and so no additional tunneling process occurs for $\omega \geq -(E_\downarrow - \mu_1)$. Thus, the steps in the noise are at $\omega = \pm(\mu_1 - E_\uparrow)$ and at $\omega = E_\downarrow - \mu_1$; see Fig. 2 (dashed

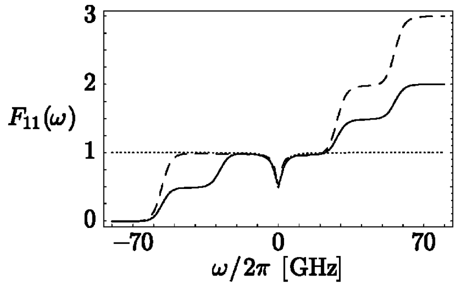

FIG. 2. The Fano factor $F_{11}(\omega) = S_{11}(\omega)/e\langle I\rangle$ in the shot noise regime $\Delta\mu > kT$ as function of noise frequency ω for a dot with two Zeeman levels. (Asymmetric noise at frequencies up to 90 GHz has been measured with a resolution compatible with this plot [8].) We consider $T = 100$ mK, $\Delta\mu/e = 460\ \mu$V, $E_\uparrow = (\mu_1 + \mu_2)/2$, $\gamma_1 = \gamma_2 = 5 \times 10^9$ s^{-1}, and $g = 2$. We use the full expression for the noise $S_{11}^{\rm b}$ [Eq. (4)] (solid line) and within Markov approximation, $S_{11}^{\rm b,Mkov}$, (dotted line), for $B = 1$ T [see Fig. 1(b)], thus $\Delta_z = \Delta\mu/4$ and $\langle I\rangle = 530$ pA. We also show $S_{ll}^{\rm c}$ (dashed line), being strongly asymmetric, where $B = 3$ T, $\Delta_z = 3\Delta\mu/4$, and $\langle I\rangle = 400$ pA. The dip near $\omega = 0$ is due to the charging effect of the dot, while the steps at ω_k (see text) arise from additional transitions for increasing ω and provide a striking effect in the quantum shot noise. Note that these steps disappear when the noise $S_{11}^{\rm b}$ is symmetrized (some features remain for $\gamma_1 \neq \gamma_2$).

line). 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 E_j gives rise to steps at $\pm(\mu_l - E_j)$ if the level is inside the bias window, $\mu_1 + kT > E_j > \mu_2 - kT$, and to a single step at $|\mu_l - E_j|$ otherwise. (More precisely, instead of the energy E_j, one has to consider the chemical potential of the dot, Δ_{bn} [19].) The height $h_\pm$ of the step at $\omega = \pm\omega_0$ is well defined for sufficiently low temperatures and is given as follows. When ω is increased above $-\omega_0 < 0$, a step arises from a tunnel transition from some initial dot state $|i\rangle$ to a final state $|f\rangle$. The step height h_- is then given by the product of the corresponding tunneling rate, γ_0, and the population of the initial state, $\rho_i = \langle i|\rho_{\rm d}|i\rangle$. The step at $\omega = \omega_0$ is determined by the reversed tunneling process, and is thus given by the product of the same tunneling rate γ_0 and the population of the final state, ρ_f. Thus, we can extract the tunneling rate from the step height and the level populations from the ratio of the step heights, $h_-/h_+ = \rho_i/\rho_f$. (If the step at $-\omega_0$ is absent, then $h_- = 0$ and thus $\rho_i = 0$; e.g., from the steps in $S_{ll}^{\rm c}$, we can extract $\bar{\rho}_\downarrow = 0$; see Fig. 2.) For sufficiently large $|\omega|$, the antisymmetric contribution becomes $\frac{1}{2}[S_{ll}(\omega) - S_{ll}(-\omega)] = \frac{1}{2}\,\mathrm{sgn}(\omega)S_{ll}(|\omega|)$, which for one or two levels is given by Eqs. (11)–(13). Finally, we stress that such a highly asymmetric $S_{ll}(\omega)$ can be observed with an appropriate measurement apparatus [2–7] and the quantum fluctuations beyond the classical limit can be accessed..

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