Shot-Noise Suppression in the Single-Electron Tunneling Regime

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Electrical current fluctuations through tunnel junctions are studied with a scanning-tunneling microscope. For single-tunnel junctions classical Poisson shot noise is observed, indicative for uncorrelated tunneling of electrons. For double-barrier tunnel junctions, formed by a nanoparticle between tip and surface, the shot noise is observed to be suppressed below the Poisson value. For strongly asymmetric junctions, where a Coulomb staircase is observed in the current-voltage characteristic, the shot-noise suppression is periodic in the applied voltage. This originates from correlations in the transfer of electrons imposed by single-electron charging effects.

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Time-dependent fluctuations in the electrical current due to the discreteness of the charge e are known as shot noise. From the shot-noise power per unit-frequency range S_I information is obtained on the conduction process which is complementary to the resistance measurement. Shot noise is well known from electron tubes and tunnel junctions, where $S_I = 2e|I| \equiv S_{\text{Poisson}}$, with I the average current. This tells us that the electrons are transmitted through the conductor as uncorrelated current pulses, i.e., as in a Poisson process.

Recently, shot noise has been investigated in mesoscopic conductors. It has been found theoretically that the shot noise can be suppressed below S_{Poisson} due to correlations in the electron transmission. There are two types of correlations: Firstly, correlations due to the Pauli exclusion principle [1,2]. This leads to a complete suppression of the shot noise in a quantum point contact, where the conductance is quantized [1], and to a suppression of one-third of S_{Poisson} in a metallic diffusive conductor [3]. Secondly, correlations can arise due to Coulomb interactions. It is well known that Coulomb interactions can have a profound effect on the current-voltage (I-U) characteristic of single-electron tunneling (SET) devices [4]. In the SET regime the capacitance of an intermediate electrode between two tunnel junctions is chosen to be so small that the addition of a single electron gives rise to a charging energy E_C much larger than the thermal energy k_BT. Hershfield *et al.* [5] and others [6] have studied theoretically the noise properties of double-barrier tunnel junctions (DBTJ's) taking into account the charging energy of the intermediate electrode. It is predicted that S_I can be suppressed below S_{Poisson}, with a suppression factor that is determined by voltage dependent tunneling rates of both barriers.

Experimentally, suppression of shot noise has been measured in quantum point contacts [7] and in diffusive conductors [8]. Li *et al.* [9] have studied shot noise in resonant tunneling diodes, where they have found S_I to vary between $\frac{1}{2}S_{\text{Poisson}}$ and S_{Poisson}, depending on the ratio of the resistances of the two barriers. Up to now no shot-noise experiments have been reported in the SET regime, where correlations due to Coulomb interactions become important.

In this Letter we report on shot-noise measurements in this regime using a scanning-tunneling microscope (STM). Previously, $1/f$ noise originating from resistance fluctuations and thermal Johnson-Nyquist noise [10] has been measured with the STM, but we do not know of any STM measurements of shot noise. For a single-tunnel junction we find that $S_I = S_{\text{Poisson}}$ indicative for uncorrelated electron tunneling. The crossover from Johnson-Nyquist to shot noise is well described by the classical formula. Subsequently, we show results of noise measurements in DBTJ's, formed by a nanoparticle between tip and surface. In this system E_C exceeds the thermal energy by 3 orders of magnitude [11]. We find S_I to be suppressed below S_{Poisson} due to correlated electron transmission induced by Coulomb interactions. The suppression depends on the two tunnel resistances and on the applied voltage.

The current noise is measured simultaneously with the current I using a series resistor R_M and a field effect transistor that are placed very close to the tunneling tip. A high frequency bandwidth of up to 200 kHz is obtained by a feedback circuit which minimizes the effective input capacitance of the transistor (see Fig. 1). Averaging the noise signal $\approx$5 s per data point, we can measure changes in the current noise as small as $\Delta S_I/2e \approx 0.04$ nA [12]. We use mechanically prepared PtRh tips and samples with epitaxially grown Au(111) films on mica substrates. Small metal particles (on average 5 nm in diameter), separated by a tunnel junction from the Au(111) surface, are formed by additionally growing a thin Zr-oxide layer, serving as a tunnel barrier, and a discontinuous gold film [11]. A DBTJ is realized by positioning the tip above such a metal particle. The STM offers the possibility to select different particles and vary the tip-particle resistance over a wide range by changing the distance z between the tip and the sample. This allows us to select the conditions for which different shot-noise behavior is expected. During the measurement

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z is kept constant, while the potential U of the sample with respect to the tip is slowly varied. The current noise is plotted as a function of the simultaneously measured current I after subtracting the preamplifier background noise, which does not depend on I.

Since generally $1/f$ noise dominates at lower frequencies, we first discuss the requirements needed to distinguish shot noise from $1/f$ noise. In STM [10], $1/f$ noise can be described by the phenomenological equation $S_I = \alpha I^2/f \equiv S_{1/f}$ [13]. The empirical parameter α depends on the system, the applied voltage, and the temperature and varies in an STM typically between 10^{-3} and 10^{-6}. The requirement that $S_{\text{Poisson}} \gg S_{1/f}$ is equivalent to $|I| \ll 2ef/\alpha$. Since our experimental results obtained at $T = 4.2$ K reported below are measured for $f = 100$ kHz with an experimentally derived parameter $\alpha \simeq 10^{-5}$, the above condition reads $|I| \ll 3$ nA. This condition is always fulfilled in our experiment where I never exceeds the value 0.5 nA. In SET devices [14] charge fluctuations produce an additional $1/f$ component, which is estimated to be below the accuracy of our shot-noise measurement at 100 kHz [15].

For arbitrary applied voltage and temperature the shot noise of a tunnel junction is given by [13]

$$S_I(T) = 2eI \coth(eU/2k_BT). \quad (1)$$

In the low voltage limit ($e|U| \ll k_BT$) Eq. (1) reduces to $S_I = 4k_BT/R$, which describes equilibrium thermal (Johnson-Nyquist) noise. For the opposite limit Poissonian shot noise is obtained. In Fig. 1 two noise measurements (triangles) are compared with Eq. (1) (solid curves) for single-tunnel junctions realized with the STM tip tunneling on an Au film (see schematics). For data (a) in Fig. 1, measured at $T = 300$ K with a tunneling resistance $R \approx 0.32$ GΩ, thermal noise dominates within the current range $|I| < 100$ pA for which $e|U| < k_BT$. On the other hand, for data (b), measured at $T = 77$ K with $R \approx 2.7$ GΩ, shot noise dominates for currents $|I| > 2.5$ pA. The experiments reported below are all done at even lower temperatures (4.2 K) and for $R \gtrsim 1$ GΩ, where the thermal noise is negligible. We would like to emphasize that, in addition to demonstrating the relation between shot and thermal noises, Fig. 1 also proves that metallic tunnel junctions of atomic dimensions, as established in the STM, exhibit full classical shot noise in the limit $e|U| \gg k_BT$.

The following experiments are obtained on samples with metallic particles. First, the STM tip is moved to a position without a particle underneath, establishing a single-tunnel junction. The corresponding noise measurement is shown in Fig. 2(a) (crosses) and serves as reference for the experiments obtained on particles. As expected, the noise corresponds to S_{Poisson} and the simultaneously measured I-U characteristic is linear (dotted line in the inset of Fig. 2). Next, the tip is positioned above a metal particle, which results in two tunnel junctions connected in series via the intermediate particle (see schematics in Fig. 2). The two junctions are characterized by the tunneling resistance R_1 and capacitance C_1 between tip and particle and by R_2 and C_2 between particle and substrate. Because of the small size of the particle, the Coulomb energy for adding one electron

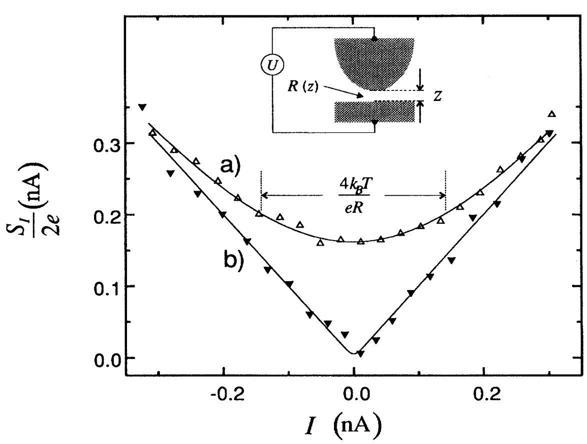

FIG. 1. Measured current noise S_I at 200 kHz vs current I, showing the transition from thermal to shot noise (a) at 300 K with $R \approx 0.32$ GΩ (open triangles), and (b) at 77 K with $R \approx 2.7$ GΩ (solid triangles). The solid theoretical curves are according to Eq. (1). Inset: schematics of the tip, sample, and the current preamplifier. Voltage fluctuations are measured across $R(z) \parallel R_M$. The transistor serves as impedance converter, and the feedback is used to minimize the effective input capacitance.

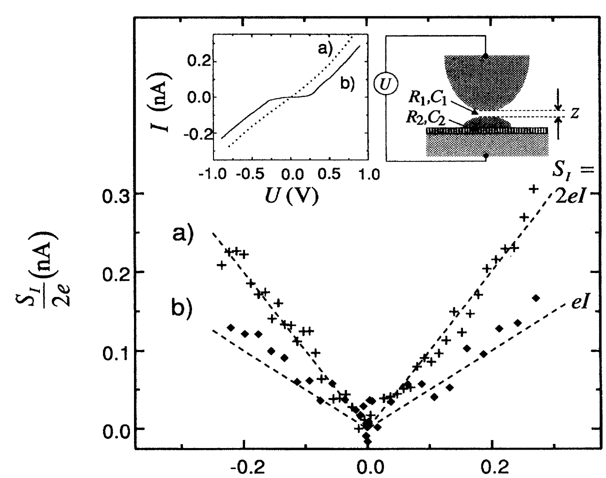

FIG. 2. Current noise S_I for (a) a single-tunnel junction (crosses) and (b) a DBTJ with $R_1 \approx 650$ MΩ and $R_2 \approx 1.4$ GΩ (diamonds), both obtained at 4.2 K. Dashed lines correspond to S_{Poisson} and $\frac{1}{2}S_{\text{Poisson}}$. Left inset: simultaneously measured I-U curve corresponding to (a) the single junction (dotted curve) and (b) the DBTJ (solid curve). Right inset: sample geometry for (b).

to the particle $E_C = e^2/2C$ ($C = C_1 + C_2$) is much larger than the thermal energy, typically $E_C \gtrsim 1000 k_B T$. Hence, SET effects are observable. The I-U curve exhibits the characteristic Coulomb blockade [11,16], a suppression of the current for $|U| \lesssim E_C/2e$ (solid curve in the inset of Fig. 2). The simultaneously measured shot noise (diamonds in Fig. 2) is suppressed relative to the single-junction data by a factor 0.59 on average. This suppression can be understood as the result of the superposition of two independent tunnel junctions with resistances R_1 and R_2 each characterized by S_{Poisson}, so that

$$S_I = S_{\mathrm{Poisson}}(R_1^2 + R_2^2)/R^2, \tag{2}$$

with $R = R_1 + R_2$ [17]. The maximum suppression of 0.5 is obtained for $R_1 = R_2$. Equation (2) is also obtained from the general shot-noise theory valid for the SET regime [5] in the asymptotical limit for $|U| \gg E_C/e$. By measuring $R = R_1(z) + R_2$ as a function of the tip-particle distance z, we obtain R_2 in the limit $z \to 0$. For data (b) in Fig. 2 we find $R_1 \approx 650\ \mathrm{M\Omega}$ and $R_2 \approx 1.4\ \mathrm{G\Omega}$, so that Eq. (2) predicts a noise suppression of 0.57, in excellent agreement with the measured suppression of 0.59. A suppression close to 0.5 has also been observed by Li *et al.* for nearly symmetric resonant-tunneling devices [9]. Though our results are obtained in the SET regime, the effects of Coulomb correlations are difficult to observe in the case of nearly symmetric junctions. This is because deviations from the classical behavior are expected to be observable only close to the Coulomb blockade, where the resulting shot noise is too small to be measured within the precision of our experiment.

For strongly asymmetric junctions ($R_1 \ll R_2$) the situation is different. Here, the I-U characteristic shows a pronounced Coulomb staircase, if simultaneously $C_1 \lesssim C_2$ holds. This condition is fulfilled because of the high dielectric constant of the oxide ($\epsilon \approx 10$) [11]. In the following $R_2 \approx 100 R_1$. The thick curve in the upper part of Fig. 3 is the measured Coulomb staircase. The thin curve shows a numerical fit obtained using $R_1/R = 0.01$, $C_1/C = 0.2$, and an offset charge [16] of $Q_0 = 0.33e$. The Coulomb staircase is well described by the "orthodox theory," which assumes inelastic scattering of the electrons on the particle [4,16]. For every step in the current the number of excess electrons on the particle increases or decreases by 1. The corresponding measured noise signal is shown by the diamonds in the lower part of Fig. 3. The solid curve has been obtained numerically following the theory of Hershfield *et al.* [5], using the same parameters as for the I-U curve with no additional fit parameters. The full shot-noise level $S_I = 2e|I|$ is periodically reached and suppressed in between, correlating with the step structure in the I-U curve. Rotating the plot 90° clockwise, one obtains the usual presentation of the Coulomb staircase. Full shot noise is obtained

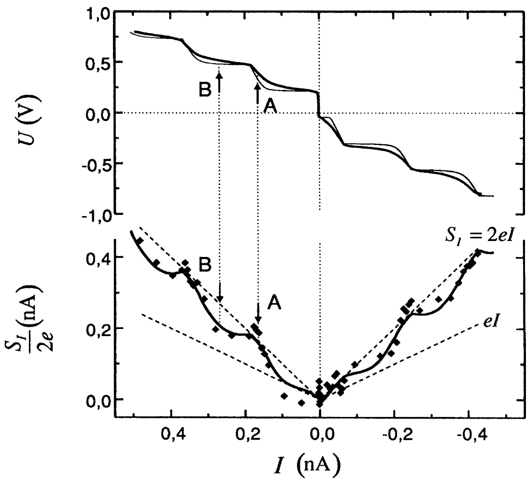

FIG. 3. Upper figure: voltage U vs current I for a DBTJ at 4.2 K, showing the Coulomb staircase. The thick curve is measured; the thin curve is numerically calculated according to [16] with $Q_0 = 0.33e$, $R_1/R = 0.01$, and $C_1/C = 0.2$. Lower figure: measured noise S_I as function of I (diamonds) and the theoretical curve (solid) calculated following the recipe given in [5], using the same parameters as above. The arrows A (B) mark positions of maximum (minimum) shot noise corresponding to a plateau (step) in the I-U curve.

for current plateaus (arrow A in Fig. 3), while the noise is suppressed for current steps in between plateaus (arrow B). In this nonclassical regime, Eq. (2), which predicts $S_I = S_{\mathrm{Poisson}}$ for $R_1 \ll R_2$, no longer applies, since the tunnel rates across both junctions depend on single-electron charging effects.

The measured steps in the I-U curve are smeared out compared to the theoretical curve (Fig. 3, upper part) due to fluctuations in Q_0 caused by the trapping and detrapping of charges in the neighborhood of the particle [18]. This limits the measuring time and thus the resolution of the noise measurement. In our experiments the voltage is typically swept during 3 s and about 60 sweeps are averaged. For the measurement shown in Fig. 4 (diamonds) we could average 300 sweeps without significant changes in the offset charge. The periodic suppression of the shot noise below the classical Poisson value is clearly observed, reaching a factor of $\frac{1}{2}$ for the position corresponding to the first step in the Coulomb staircase. This is in excellent agreement with theory (solid curve in Fig. 4) [5].

An explanation for the shot-noise suppression caused by charging effects can be given by the two-state model [5]. Only the two charge states with the energetically most probable number of electrons on the particle are considered, which is a valid approximation for $E_C \gg k_B T$ and $U \approx e/C$. Within this model tunneling always occurs alternately across the two junctions. After an electron has been added to the particle by tunneling across junction 1, this junction is blocked until the

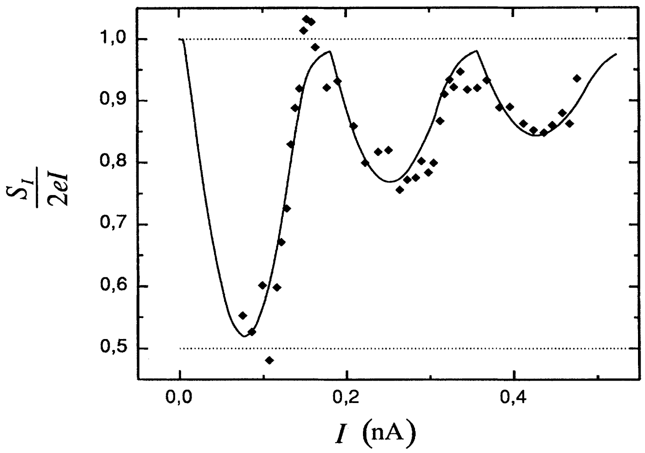

FIG. 4. Ratio of the measured current noise S_I to S_{Poisson} as a function of current I (diamonds). The solid theoretical curve is calculated for $T = 4.2$ K, $Q_0 = 0.33\,e$, $R_1/R = 0.01$, and $C_1/C = 0.2$ [5].

electron is removed by tunneling across junction 2. On a current plateau in the Coulomb staircase the number of excess electrons is constant for most of the time, only disturbed during a very short instant. In case $R_2 \gg R_1$, the tunneling events are solely determined by junction 2, hence the shot noise is as of a single-tunnel junction. The situation changes completely for voltages where the Coulomb staircase shows a step. Here the two charge states are degenerate in total energy, which results in similar tunneling probabilities for the two junctions. As both junctions are on average alternately blocked during *equal times*, tunneling becomes correlated causing a suppression of shot noise with a maximum of $\frac{1}{2}$.

In conclusion, we have studied shot noise in single-tunnel junctions and double-barrier tunnel junctions operated in the single-electron tunneling regime using an STM. We have observed shot-noise suppression in DBTJ's, which depends on the resistance ratio R_1/R_2 and the applied voltage. For comparable tunnel resistances the suppression is given by the factor $(R_1^2 + R_2^2)/R^2$, whereas for strongly asymmetric tunneling resistances charging effects lead to a voltage dependent correlation between the tunneling events. The shot-noise suppression is observed to be periodic, with a maximum suppression factor of $\frac{1}{2}$.

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