Crossover between classical and quantum shot noise in chaotic cavities

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The discreteness of charge in units of *e* led Schottky in 1918 to predict that the electrical current in a vacuum tube fluctuates even if all spurious noise sources are eliminated carefully[1]. This phenomenon is now widely known as shot noise. In recent years, shot noise in mesoscopic conductors, where charge motion is quantum-coherent over distances comparable to the system size, has been studied extensively[2–5]. In those experiments, charge does not propagate as an isolated entity through free space, as for vacuum tubes, but is part of a degenerate and quantum-coherent Fermi sea of charges. It has been predicted that shot noise in mesoscopic conductors can disappear altogether when the system is tuned to a regime where electron motion becomes classically chaotic[6]. Here we experimentally verify this prediction by using chaotic cavities where the time that electrons dwell inside can be tuned[7]. Shot noise is present for large dwell times, where the electron motion through the cavity is 'smeared' by quantum scattering, and it disappears for short dwell times, when the motion becomes classically deterministic.

Noise is generally due to randomness, which can be classical or quantum in nature. Randomness can be inherent in the reservoirs emitting charges or in the transmission process between emitter and collector. Shot noise observed in vacuum tubes[8,9] (Fig. 1a) is solely due to fluctuations of the state occupancy in the reservoirs, leading to the completely random emission of electrons into vacuum. This random emission results in a spectral density S of the current fluctuations given by Schottky's formula $S = 2e|I|$ (I denotes the mean electrical current). But the transmission from cathode (K in Fig. 1a) to anode (A) is free of randomness, because an emitted electron will eventually reach the anode with absolute certainty. Loosely speaking, the vacuum is noiseless.

In a coherent electrical conductor at zero temperature, the situation is the other way around (Fig. 1b). The emission from an ideal reservoir (a Fermi gas) into the wire is noiseless[10]. Here, randomness is caused by scattering within the conductor, which can be either classical (deterministic) or quantum-mechanical. Both add to the electrical resistance, but only quantum scattering generates noise[11]. This fundamental statement is verified experimentally in the present work by measuring the shot noise of open chaotic cavities.

Figure 2a shows a schematic drawing of a chaotic cavity connected via two narrow constrictions to a left (L) and a right (R) reservoir. At the contacts, charge transport takes place within an integer number N of energy channels, each with transmission probability $T = 1$. As electrons pass the contacts with unit probability, they are noiseless. Nevertheless, there is a fundamental resistance $R_{L,R}$ associated with each contact: $R_{L,R}^{-1} = G_{L,R} = (2e^2/h)N_{L,R}$ (refs 12 and 13). Inside the cavity the electron motion is chaotic. An electron entering the cavity from the left contact scatters inside the cavity and leaves it after the dwell time τ_D either at the left or the right contact with probabilities determined by the conductances $G_{L,R}$. The randomization inside the cavity leads to a total resistance R which is simply the sum of the left and right contact resistances: $R = R_L + R_R$ (the region inside the cavity has negligible resistance). Note that $R = R_L + R_R$ holds irrespective of whether chaos within the cavity is classical or quantum. As we will show below, this is markedly different for shot noise, the occurrence of which relies on the presence of quantum scattering—that is, diffraction caused by the wave nature of electrons. Because the degree of diffraction can be tuned in the experiment by changing the effective dwell time τ_D, this statement can experimentally be verified.

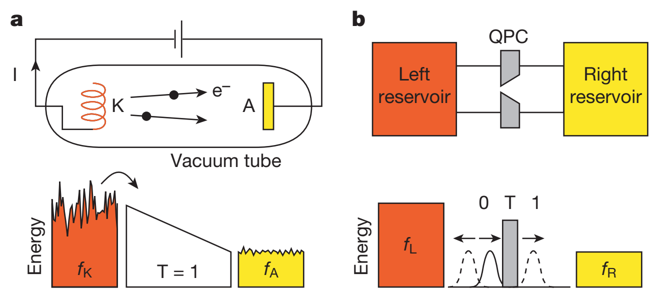

Figure 1 Origin of noise. **a**, In a classical vacuum tube, the randomness that gives rise to shot noise stems solely from fluctuations in the reservoirs. The transmission from cathode (K) to anode (A) through the vacuum is noiseless, because it occurs with unit probability. **b**, A quantum point contact (QPC)[12,13] as a coherent conductor: at zero temperature randomness is due to scattering within the conductor only. Although this scattering can generally be classical or quantum-mechanical, only quantum scattering generates noise. (Here f denotes the distribution of electrons within the respective reservoirs and T the transmission probability.)

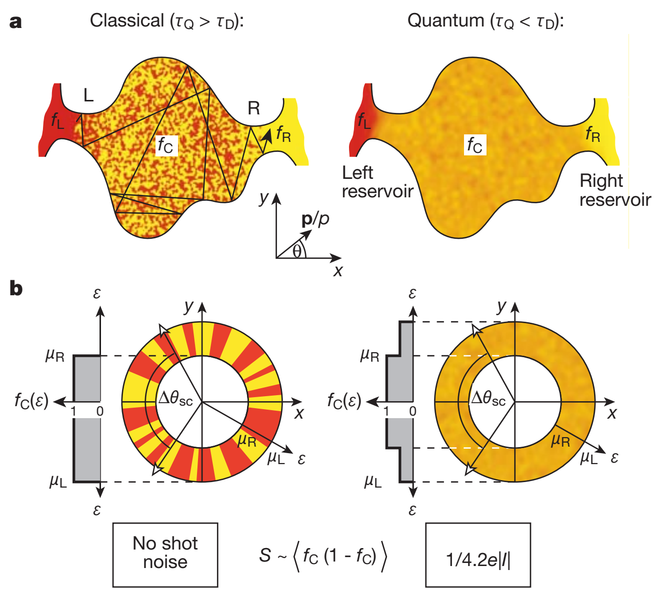

Figure 2 Chaotic cavity in the classical and quantum regime. **a**, Diagrams of a chaotic cavity connected by two contacts to a left (L) and right (R) reservoir. In the classical regime of deterministic scattering, the electrons with a specific momentum direction $\mathbf{p}/p$ inside the cavity either originate from the left (red) or the right (yellow) reservoir. The chaotic motion leads to a distribution function f_C inside the cavity that randomly switches between 0 (yellow) and 1 (red) within the relevant energy interval $[\mu_R, \mu_L]$. This is illustrated by the speckle pattern within the cavity. In the quantum case, the wavefunction of the electrons is spread over the whole cavity and we are not able to decide whether an electron stems from the left or the right side. **b**, The shot noise power is determined by the fluctuations $\langle f_C(1 - f_C)\rangle$ of the state occupancy f_C. Because classically f_C takes on either the value 0 or 1, the noise $S \propto \langle f_C(1 - f_C)\rangle$ is zero in this case, although $\langle f_C \rangle \neq 0, 1$. If quantum diffraction occurs, f_C can take on an arbitrary value between 0 and 1 within the energy interval $[\mu_R, \mu_L]$. In the 'full' quantum limit (strong diffraction) $\langle f_C(1 - f_C)\rangle = \langle f_C \rangle(1 - \langle f_C \rangle)$ leading to a shot noise of $(1/4)2e|I|$ for a symmetric cavity, because $\langle f_C \rangle = 1/2$ in this case.

The shot noise power of an open cavity can be expressed in terms of the distribution function f_C of the electrons inside the cavity[14]:

$$S = (2/R)\int d\epsilon \langle f_C(\mathbf{p},\mathbf{r})[1 - f_C(\mathbf{p},\mathbf{r})]\rangle \qquad (1)$$

Here $\langle\cdots\rangle$ denotes the average over momentum direction $\mathbf{p}/p$ and spatial coordinate $\mathbf{r}$, and ε denotes the energy. There are three important timescales in the problem: (1) the ballistic flight time τ_F, which is the time an electron takes to traverse the cavity once; (2) the dwell time τ_D, and (3), the quantum scattering time τ_Q, which qualitatively is the mean time during which the classical trajectory is 'lost' by diffraction. Because of the relatively large cavities and small openings in the present experiments, $\tau_D \gg \tau_F$.

We first look at the classical regime, characterized by $\tau_Q \gg \tau_D$. In this regime classical (deterministic) trajectories are well defined. The distribution function f_C at a given point within the cavity and for a given energy ϵ can be expressed as a function of momentum direction $\mathbf{p}/p$ (Fig. 2). As the cavity is chaotic, a large number of different trajectories cross this given point. However, because classical physics is deterministic, each trajectory can be traced back to its unique origin, which is either the left or the right contact. Hence f_C is either one or zero. The product $f_C(1 - f_C)$, therefore, always equals zero and shot noise is absent. Intuitively, this appears to be very surprising, because of the presence of chaos. Classical chaos leads to a strange function f_C, which only takes on the values 0 and 1, but may switch between these two values in a very erratic and dense way (Fig. 2b). This, however, does not produce noise.

If, on the other hand, quantum diffraction cannot be neglected, that is, $\tau_Q < \tau_D$, the situation is very much changed. Owing to quantum scattering on impurities and at the boundary of the cavity, an electron wave (classically the 'trajectory') may split into two or more partial waves leaving the cavity at different exits. A momentum state within the cavity cannot be traced back unambiguously to either contact, but carries information of both contacts simultaneously. Consequently, f_C is a weighted sum of f_L and f_R and can now take on values between 0 and 1. This uncertainty of not knowing where the electron came from and where it will go to is the source of noise.

The crossover between classical and quantum regimes in chaotic cavities has been theoretically discussed[6]. For a symmetric cavity, the noise power S is given by:

$$S = S_{eq}[1 + F(\beta \coth\beta - 1)], \quad \beta = \frac{eV}{2k_B\Theta} \qquad (2)$$

where $F \equiv S/2e|I|$ is the Fano factor defined as the current-normalized noise power at zero temperature, and $S_{eq} = 4k_B\Theta/R$ the equilibrium (thermal) noise of the contacts.. V denotes the applied voltage, and Θ the temperature. The Fano factor F decays to zero in the classical limit where $\tau_Q/\tau_D \to \infty$ and is to first order in τ_Q/τ_D given by:

$$F = \frac{1}{4}\left(1 - \frac{\tau_Q}{\tau_D}\right) \qquad (3)$$

The noise power S can be written as $S = S_{CL} + S_Q$, where $S_{CL} = S_{eq}$. This part is purely classical. In contrast, the second non-equilibrium part $S_Q \propto F$ is a sensitive probe of quantum mechanics.

Cavities of size L comparable to the mean free path l are experimentally realized in a two-dimensional electron gas (Fig. 3a, right inset). Two quantum point contacts, electrostatically defined by metallic split gates, connect the cavity to the reservoirs. On the lateral two sides the electrons are confined by wet chemical etching of the two-dimensional electron gas. The openings of the cavity can be altered by varying the gate voltages. So-called 'open' cavities are defined when both quantum point contacts are adjusted

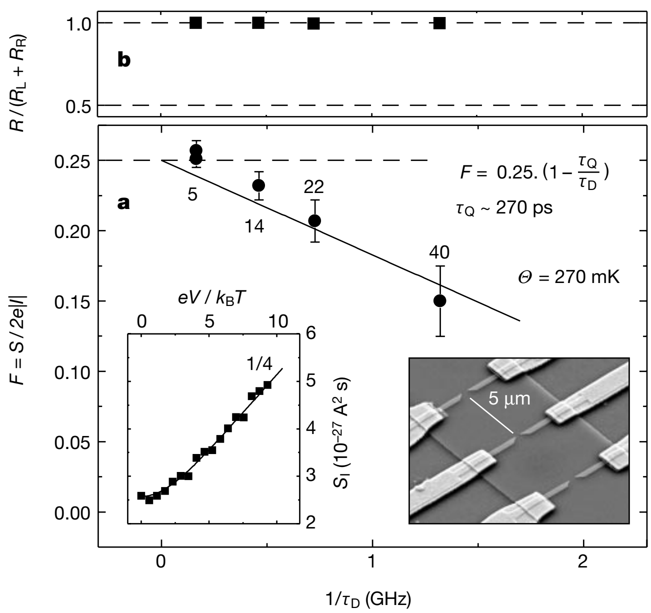

Figure 3 Fano factor $F \equiv S/2e|I|$ versus inverse dwell time τ_D^{-1} of a symmetric cavity ($N_L = N_R$). **a**, With increasing τ_D^{-1}, F becomes smaller than 1/4 (quantum limit), indicating a crossover from full quantum to deterministic (classical) scattering inside the cavity. The numbers at the data points indicate the number of fully transmitted modes N at the contacts. **b**, Measured ratio between the total resistance R and the series resistance $R_L + R_R$ of the two contacts as a function of τ_D^{-1}. As the ratio $R/(R_L + R_R)$ is always 1, the electronic motion inside the cavity is chaotic. Left inset, shot noise for $N = 5$ in the full quantum regime, resulting in a Fano factor of 1/4. Right inset, scanning electron microscope picture of the cavity.

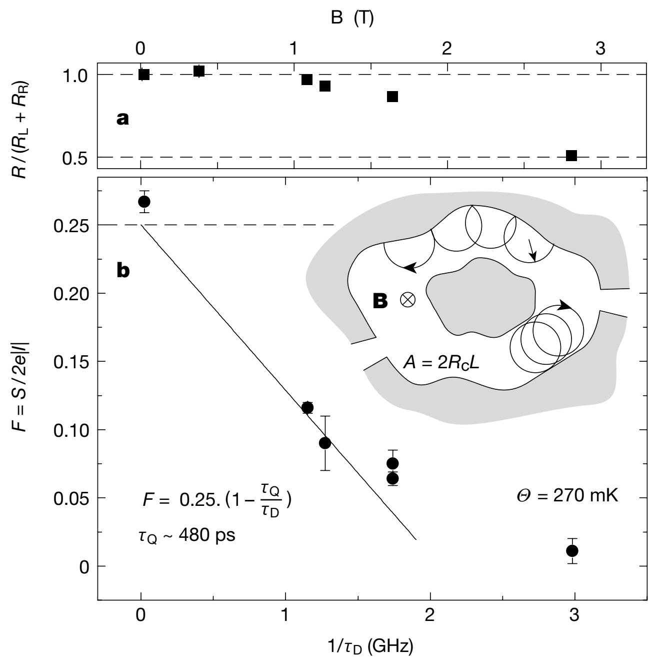

Figure 4 Measured Fano factors in various magnetic fields. A magnetic field reduces the dwell time: $\tau_D^{-1} \propto B$. Theoretically, this effect can be described as a change of the area of the cavity, because for fields corresponding to a cyclotron radius R_C smaller than the size L of the cavity, an annulus is formed at the edge with an inner part of the cavity not contributing to transport and noise (see inset). **a**, Total resistance R normalized to the series resistance $R_L + R_R$ as a function of the inverse dwell time τ_D^{-1}. **b**, F deviates from the quantum limit 1/4 with increasing τ_D^{-1}, indicating a crossover from full quantum to classical scattering inside the cavity similar to the measurements in zero magnetic field (Fig. 3a).

to a conductance plateau with an integer number N of fully transmissive ($T = 1$) modes. Experimental details are given in ref. 7.

The deviations from the quantum limit[15] $F = 1/4$ towards the regime of deterministic scattering $F < 1/4$, described by equation (3), are explored by changing τ_D. This dwell time depends on the area A of the cavity and the conductances $G_{L,R}$ of the contacts[7]:

$$\tau_D = \frac{2e^2 m}{\pi \hbar^2} \frac{A}{(G_L + G_R)} \quad (4)$$

In a first experiment we changed the openings (conductances) of the contacts to alter τ_D. In Fig. 3a, the Fano factor F of a symmetric ($N_L = N_R$) cavity is plotted as a function of the inverse dwell time τ_D^{-1} for four different settings ($N_L = N_R = 5, 14, 22$ and 40). The left inset shows the measured shot noise for $N_L = N_R = 5$. As the contacts were further opened and the dwell time was subsequently reduced, the shot noise was observed to decrease. The Fano factor F shows a pronounced decay below the quantum limit 1/4. A linear fit of the data to equation (3) yields a τ_Q of 270 ps. We emphasize that the total resistance R equals the series resistance $R_L + R_R$ of the two contacts within the measurement accuracy of ~3% (Fig. 3b). Hence, the shot noise measurements are carried out in a regime where the direct (ballistic) transmission of electrons from the left to the right contact can be neglected. The suppression of shot noise observed here is a consequence of reduced diffraction, and serves to demonstrate that shot noise disappears in the limit of purely classical scattering.

An alternative way to change τ_D is to apply a perpendicular magnetic field. Because a magnetic field forces the electrons onto circular orbits with the cyclotron radius $R_c = mv_F/eB$ (where m denotes the electron mass and v_F the Fermi velocity), the dwell time τ_D will be reduced with increasing magnetic field B provided that $R_c < L$. An annulus of skipping orbits is formed (Fig. 4b inset) in which transport takes place. Such an annulus represents a 'new' (and smaller) chaotic cavity inside the actual cavity. For low magnetic fields (large filling factors), the electron dynamics inside the annulus can still be considered to be random (because of impurities or irregularities in the geometry of the cavity). Thus equations (2) and (3) are still valid with the area A in equation (4) replaced by $A = 2R_cL_c$, where $L_c \approx L$ is the circumference of the cavity. This leads to $\tau_D^{-1} \propto B$.

Figure 4b shows the measured Fano factor F as a function of inverse dwell time τ_D^{-1} in a magnetic field. We again observe a very marked reduction of F with increasing τ_D^{-1}, while the total resistance R approximately equals the series resistance $R_L + R_R$ of the two contacts in lower fields ($B < 1.2$ T) (Fig. 4a). A linear fit (equation (3)) results in a quantum scattering time τ_Q of ≈485 ps, which is in qualitative agreement with τ_Q obtained from the measurements in zero field. If the magnetic field is further increased beyond 1.2 T, the ratio $R/(R_L + R_R)$ starts to deviate from unity and equals 1/2 at the highest magnetic field. Here, we enter a new regime in which a significant fraction of electrons is ballistically transmitted from source to drain. The last measurement point in Fig. 4 with $F = 0$ and $R/(R_L + R_R) \approx 0.5$ corresponds to the integer quantum Hall regime with filling factor four, where the electrons propagate within ballistic edge states. □

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Observation of stimulated emission by direct three-photon excitation

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Multiphoton processes, predicted[1] theoretically in 1931, were for a long time considered to be mainly of academic interest. This view changed when it was shown[2,3] that a two-photon absorption process could, because of a quadratic dependence of excitation on intensity, produce a spatially confined excitation useful for three-dimensional data storage and imaging. Two-photon absorption has received considerable attention recently because of the development of highly efficient two-photon-sensitive materials, leading to numerous technological applications[4–28]. These successes have created interest in exploring applications based on three-photon excitations[29]. For a three-photon process, a longer excitation wavelength such as those common in optical communications can be used. Also, the cubic dependence of the three-photon process on the input light intensity provides a stronger spatial confinement, so that a higher contrast in imaging can be obtained. Here we report the observation of a highly directional and up-converted stimulated emission as an amplified spontaneous emission, produced in an organic chromophore solution by a strong simultaneous three-photon absorption at 1.3 μm. This achievement suggests opportunities for a three-photon process in frequency-upconversion lasing, short-pulse optical communications, and the emerging field of biophotonics.

In our experiment, the gain medium is the organic chromophore 4-[N-(2-hydroxyethyl)-N-(methyl)amino phenyl]-4′-(6-hydroxyhexyl sulphonyl) stilbene (APSS) dissolved in dimethyl sulphoxide (DMSO). APSS has been reported[12] to exhibit good properties for two-photon pumped lasing as well as for optical power limiting, when excited at a wavelength of ~800 nm. The molecular structure of APSS and its linear absorption spectrum in DMSO were shown in our previous publications[12]. We report here that APSS also exhibits a strong three-photon absorption when excited by ultrashort laser pulses at ~1.3 μm, producing population inversion. This creates the prospect of developing new three-photon materials with even more enhanced cross-sections for three-photon absorption. Our