Supporting Information

Anomalous Non-linear Shot Noise at High Voltage Bias

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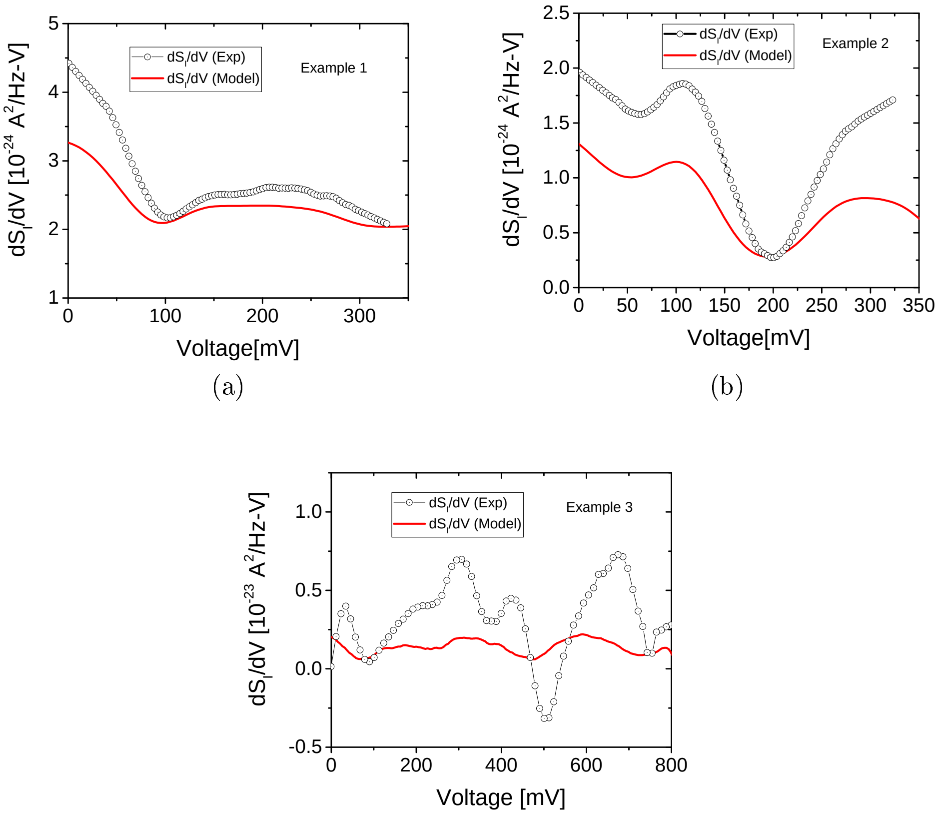
Defects on both sides of the point contact

If we include scattering over defects present both before (left) and after (right) the point contact, the expression for the transmission $T(E,V)$ will be:

$$T(E,V) = T_0 + \sum_{p=0}^{N_r} a_p \sin\left(2L_p \frac{\sqrt{2m(E+eV/2)}}{\hbar} + \phi_p\right) + \sum_{q=0}^{N_l} A_q \sin\left(2\mathcal{L}_q \frac{\sqrt{2m(E-eV/2)}}{\hbar} + \Phi_q\right) \tag{1}$$

Where N_l and N_r are the numbers of defects on the left and right of the point contact, respectively. As long as $eV \ll E_{\mathrm{F}}$ the sine terms in the summation for the left and right defects have the same dependence on V, so a defect sitting on the left will give the same frequency dependence (within a constant phase factor) as a defect at the same radial distance on the right. So, in our model, we used the expression with defects sitting only on one side of the point contact. Depending on whether defects sit on the left or right side of the point contact, the differential conductance depends only on the either T_{H} or T_{L}, as demonstrated in the main text. When a generalised expression for transmission as given above is used, with defects sitting on both the sides, then the differential conductance depends on both T_{H} and T_{L}.

Noise derivative comparison



(c)

Figure S1: Comparison of the numerical derivative of the experimental noise data and $\mathrm{dS}_I/\mathrm{dV}$ derived from the quantum interference model described in the main text for (a) Ex1, (b) Ex2 and(c) Ex3 discussed in the main text. The curves are offset for clarity.

Two-channel fit

We will include here a second constant transmission channel to our model described in main text to explain the nonlinear shot noise data shown in Figure 3. The transmission value for this second channel is extracted from the experimental dataset. For this we need to solve a set of equations for the low bias noise and differential conductance:

$$S_{\mathrm{I}} = 4\frac{e^3}{h}V\left[T_1(1-T_1)+T_2(1-T_2)\right] \tag{2}$$

Next, one can input T_2 in terms of T_1 using Landauer's conductance formula i. e. $T_2 = G_0 - T_1$, here $G_0 = G(0V)$. This gives:

$$\frac{S_{\mathrm{I}}}{V} = 4\frac{e^3}{h}\left[T_1(1-T_1)+(G_0-T_1)(1-G_0+T_1)\right] \tag{3}$$

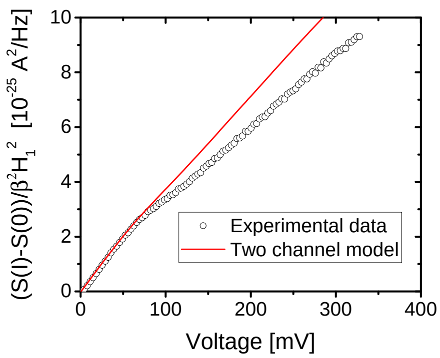
In the above equation the left hand side is the slope of measured experimental noise close to zero bias. This can be obtained from experiment. Thus we are left with a quadratic equation in T_1. On solving this we can get the zero bias transmission for the two channels involved in transport i. e. T_1 and T_2.

However, we don't know how the transmission of two channels will evolve at higher bias. Therefore, we make an assumption that the second channel transmission remains constant at all bias and the first channel transmission is responsible for the measured differential conductance shifted down by a value equal to the transmission of second channel. This constant transmission assumption adds to the noise calculated by the model, but it smooths out the non-linearity as shown in Figure S2. The reduced non-linearity obtained by adding a constant second channel demonstrates that the assumption of this second channel being constant is likely invalid. Also qualitatively we can see that the non-linearities would be enhance when the second channel transmission decreases at high bias, as suggested by the calculations by Brandbyge *et al.* [1]

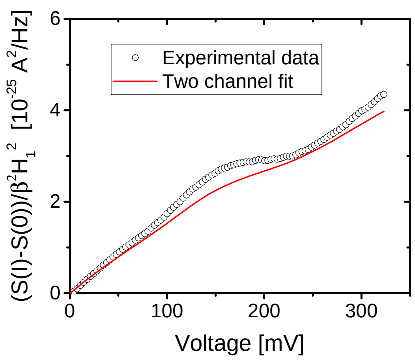
Making the complementary assumption (i. e. constant large transmittance first channel and taking the non-linearity originating only from the smaller transmittance second channel) will give the same result. The way we can see this is through equation 4 in main text. This equation is for a single channel transmission. In a two channel case where one channel is constant, the same equation can be used with the T_0 term now incorporates constant part of both the channels while the sine terms in the summation provide together the non-linear part. Now, irrespective of whether we assign this non-linear part to first or second channel, we will get the same result for noise. However, it is important to keep in mind that the transmission of both channels should lie in the range [0,1].

References

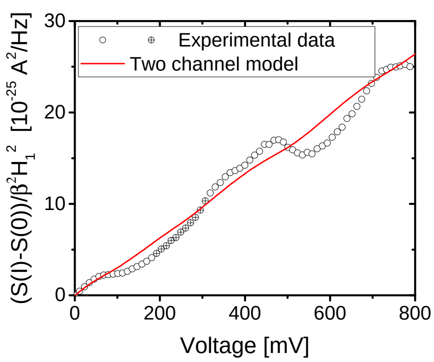
(1) Brandbyge, M.; Kobayashi, N.; Tsukada, M. Conduction channels at finite bias in single-atom gold contacts. *Phys. Rev. B* **1999**, *60*, 17064–17070.



(a)



(b)



(c)

Figure S2: Two channel fit after assuming a constant transmission contribution from the second channel for (a) Ex1, (b) Ex2 and(c) Ex3. The points for which the noise spectra show non-white character are shown with filled circles.