Shot Noise in Nanoscale Conductors From First Principles

Yu-Chang Chen and Massimiliano Di Ventra

Department of Physics, Virginia Polytechnic Institute and State University, Blacksburg, Virginia 24061

We describe a field-theoretic approach to calculate quantum shot noise in nanoscale conductors from first principles. Our starting point is the second-quantization field operator to calculate shot noise in terms of single quasi-particle wavefunctions obtained self-consistently within density functional theory. The approach is valid in both linear and nonlinear response and is particularly suitable in studying shot noise in atomic-scale conductors. As an example we study shot noise in Si atomic wires between metal electrodes. We find that shot noise is strongly nonlinear as a function of bias and it is enhanced for one- and two-Si wires due to the large contribution from the metal electrodes. For longer wires it shows an oscillatory behavior for even and odd number of atoms with opposite trend with respect to the conductance, indicating that current fluctuations persist with increasing wire length.

The subject of steady-state current fluctuations (shot noise) has attracted much attention from both theorists and experimentalists over the past decade. Shot noise is due to charge quantization, and, as such, is generally unavoidable even at zero temperature. While in signal processing shot noise might be considered as an undesirable effect that blurs the signal detection, in mesoscopic and nanoscale systems it is a physical mechanism that can provide useful information to probe the electron energy distribution, the kinetics of electrons, and electron interactions due to the Coulomb repulsion and the Pauli exclusion principle.

When electrons in a conductor diffuse in a completely uncorrelated way, the shot noise magnitude reaches the well known Poisson limit, where is the electron charge and is the dc current. In all other cases, shot noise is proportional to the average current times a real number , called Fano factor. Except for some special cases, like, e.g., transport in resonant-tunneling diodes, where shot noise is significantly enhanced in the negative differential resistance region, due to enhanced electron interaction in the well, the Fano factor is generally lower than one. In a two-terminal conductor with channels, the Fano factor can be expressed in linear response and at zero temperature in terms of the transmission coefficients of each channel

\[
F = \frac{\sum_n T_n (1 - T_n)}{\sum_n T_n},
\]

where the denominator is simply proportional to the average current. In principle, if the transmission probabilities are known, shot noise can be evaluated using Eq. (1). However, the transmission probabilities, and consequently shot noise, are an interrelated function of the electronic and ionic distributions which are generally nonlinear in the external bias. These distributions need to be calculated self-consistently at all external voltages, and can not be simply derived from ground state calculations. The last point is particularly relevant in nanoscale conductors where the chemistry of single atoms is extremely important in determining the current-voltage characteristics of the whole sample.

In this Letter we describe a field-theoretic approach to calculate quantum shot noise in nanoscale conductors where the electronic and ionic distributions are calculated self-consistently. In particular, we derive an expression of shot noise in terms of single-particle wavefunctions. The latter quantities can then be determined using a scattering approach within the density-functional theory of many-electron systems. As an example we apply the method to the study of shot noise in Si atomic wires between metal electrodes. We find that shot noise is strongly nonlinear as a function of bias and it is enhanced for very short wires due to the large contribution from the metal electrodes, while for longer wires it shows an oscillatory behavior for even and odd number of atoms.

Let us start by writing the field operator of propagating electrons for a sample connected to a left (L) and right (R) reservoir in terms of single-particle wavefunctions \( \Psi_E^{L(R)}(r, K) \) with energy \( E \) and component of the momentum parallel to the electrode surface \( K \). (see Fig. 1 for a schematic of the system investigated)

\[
\Psi = \Psi^L + \Psi^R,
\]

where

\[
\Psi^{L(R)} = \sum_E a^{L(R)}_E(t) \Psi^{L(R)}_E(r, K).
\]

The coefficients \( a^{L(R)}_E(t) = \exp(-i\omega t) a^{L(R)}_E \) are the annihilation operators for electrons incident from the left (right) reservoir, satisfying the usual anticommutation relations: \( \{ a_i^\dagger, a_j \} = \delta_{ij} \delta(E - E') \) We assume that the electrons coming from the left (right) electrodes thermalize completely in the reservoirs, i.e., the statistics of electrons coming from the left (right) electrodes is determined by the equilibrium Fermi-Dirac distribution function \( f^L(R)_E \) in the left (right) reservoir, i.e.,

\[
< a_i^\dagger a_j^\dagger > = \delta_{ij} \delta(E - E') f^L(R)_E,
\]

with i, j=R, L.

From the above field operator we can define the current operator

\[
\hat{I}(z, t) = -i \int dR \int dK \left( \Psi^\dagger \partial_z \Psi - \partial_z \Psi^\dagger \Psi \right),
\]

where
where $d\mathbf{R}$ defines an element of the electrode surface. For sake of simplicity we will assume in the following that the right Fermi level $E_{FR}$ is higher than the left Fermi level $E_{FL}$, so that the average value of the current at zero temperature is simply

$$< \hat{I} > = - i \int_{E_{FL}}^{E_{FR}} dE \int d\mathbf{R} \int d\mathbf{R}' \left( \hat{I}_{E,E}^{R,R} \right),$$

(6)

where

$$\hat{I}_{E,E'}^{R,R} = (\Psi_{E}^{\dagger})^* \nabla \Psi_{E'} - (\Psi_{E'}^{\dagger})^* \Psi_{E} ,$$

(7)

with $i,j=R,L$.

We now calculate the shot noise as the Fourier transform of the electric current auto-correlation function in the limit of zero frequency and zero temperature. [23] The electric current we refer to is the excess current $\Delta \hat{I}$ with respect to the average current Eq. (6). We first consider the spectral density

$$2\pi S(\omega) = \int dt e^{i\omega t} \left\langle \Delta \hat{I}(t) \Delta \hat{I}(0) \right\rangle ,$$

(8)

where $\Delta \hat{I}(z,t) = \hat{I}(z,t) - < \hat{I} >$ corresponds to the excess (with respect to the average) current operator. We evaluate the quantum statistical expectation value in Eq. (8) by using the Bloch-De Dominics theorem [23] (see also Ref. [16]) to obtain

$$S(\omega) = \sum_{i,j=L,R} \int dE \int_{E_{FL}}^{E_{FR}} \left( 1 - f_{E}^{i} \right) \int d\mathbf{R}_{1} \int d\mathbf{K}_{1} \int_{E_{FL}}^{E_{FR}} \int d\mathbf{R}_{2} \int d\mathbf{K}_{2} \int_{E_{FL}}^{E_{FR}} \Delta \hat{I}_{E_{FL}}^{ij},$$

(9)

In the limit of zero frequency and zero temperature the Fermi distribution function reduces to the step function $f_{E} = \theta (E_{FL,R} - E)$, and the noise power $2\pi S(\omega = 0) = \lim_{T \to 0} \int dt \left\langle \Delta \hat{I}(t) \Delta \hat{I}(0) \right\rangle$ can be written as

$$S = \int_{E_{FL}}^{E_{FR}} dE \int d\mathbf{R} \int d\mathbf{K}_{1} \int_{E_{FL}}^{E_{FR}} \left| \hat{I}_{E,E}^{R,R} \right|^{2},$$

(10)

where the range of energy integration is from $E_{FL}$ to $E_{FR}$ and $\hat{I}_{E,E}^{R,R}$ has been defined in Eq. (7).

Equation (10) is the desired expression that relates current fluctuations to single-particle wavefunctions. As it was previously noted, it is clear from Eq. (10) that shot noise is not simply proportional to the conductance of the sample [24] but is determined by an interplay between electron wavefunctions incident from the left electrode and electron wavefunctions incident from the right electrode. [24] It is also interesting to point out that at zero temperature and for a finite bias, there are no electrons incident from the left electrode in the energy region between left and right Fermi levels (see Fig. 1b). The nonzero value of $S$ is thus a direct consequence of quantum mechanical statistics reflecting the fact that current fluctuations at zero temperature are a purely quantum mechanical effect.

Equation (10) can be applied to study the behavior of shot noise in any system once the single-particle wavefunctions are known. We evaluate these wavefunctions self-consistently using the Lippmann-Schwinger equation [23]

$$\Psi_{E}^{L(R)}(\mathbf{r}, \mathbf{K}_{||}) = \Psi_{0,E,\mathbf{K}_{||}}^{L(R)}(\mathbf{r}) + \int d\mathbf{r}_{1} \int d\mathbf{r}_{2} \int \left( \mathbf{r}_{1}, \mathbf{r}_{2} \right) \Psi_{E}^{L(R)}(\mathbf{r}_{2}, \mathbf{K}_{||}) V(\mathbf{r}_{1}, \mathbf{r}_{2}),$$

(11)

where $\Psi_{0,E,\mathbf{K}_{||}}^{L(R)}(\mathbf{r}) = \exp \left( i \mathbf{K}_{||} \cdot \mathbf{r} \right) u_{E,\mathbf{K}_{||}}^{L(R)}(z)$ are the wavefunctions corresponding to propagating electrons incident from the left (right) electrode in the absence of the scattering potential $V(\mathbf{r}_{1}, \mathbf{r}_{2})$, $G$ is the corresponding Green’s function, and $\mathbf{R}$ and $\mathbf{z}$ are the coordinates parallel and perpendicular to the electrode surface, respectively. [23] Deep into the electrodes ($z \to \pm \infty$), the wavefunctions $u_{E,\mathbf{K}_{||}}^{L(R)}(z)$ assume different scattering boundary conditions according to their energy and parallel momentum. [23] The potential $V(\mathbf{r}_{1}, \mathbf{r}_{2})$ describes the difference in potential between the complete system and the bare electrodes. [23] It is the sum of the nuclear, Hartree and exchange-correlation potentials. [23] We choose to represent the latter in the local-density approximation to density-functional theory. [23] Once the single-particle wavefunctions are calculated self-consistently using Eq. (11), the electric current and shot noise can be calculated using Eqs. (8) and (10). Current-induced forces and, consequently, the ionic distribution in steady state can also be determined with the above wavefunctions. [23]

The current operator of interest to us is the one corresponding to the extra current due to the presence of

\[ \hat{I} = \sum_{i,j=L,R} \int dE f_{E}^{i} \int d\mathbf{R} \int d\mathbf{K} \int_{E_{FL}}^{E_{FR}} \Delta \hat{I}_{E,E}^{i,j}, \]

\[ \int d\mathbf{R}_{1} \int d\mathbf{K}_{1} \int_{E_{FL}}^{E_{FR}} \int d\mathbf{R}_{2} \int d\mathbf{K}_{2} \int_{E_{FL}}^{E_{FR}} \Delta \hat{I}_{E_{FL}}^{ij}, \]
The bulk electrodes are modelled with (see Fig. 1) as a function of the wire length and as a noise for Si atomic wires between two bulk electrodes when the spacing between the silicon atoms is fixed at 4.2 a.u., and the distance between the electrode edge and the first silicon atom is 2.1 a.u., the relaxed atomic positions at zero bias. We keep the ionic configuration of the system unchanged at all different biases. In Fig. 2 we plot the Fano factor (defined as $F = S/2eI$) as a function of the number of Si atoms for an external bias of 0.01 V (linear response regime). In the same plot (right axis) we also show the conductance in units of the quantum of conductance $\frac{2e^2}{h}$. The conductance of the wires oscillates as a function of the number of atoms. The origin of even and odd parity oscillations is due to the fact that even-atom-number wires have fully occupied $\pi$ orbitals, while the odd-atom-number wires have a half-filled $\pi$ state at the Fermi level. This behavior has also been predicted for C-atom wires and can be similarly explained in terms of lower (larger) density of states at the Fermi level of the total system (electrodes plus atoms) for even (odd) number of atoms. This is shown in Fig. 3 where the density of states at the left Fermi level (the right Fermi level is at 0.01 eV above the left Fermi level) is plotted as a function of the number of atoms. The Fano factor follows a similar oscillatory pattern. It is lower (larger) for odd (even) number of atoms, exactly opposite to the conductance oscillations behavior. This result can be rationalized by noting that lower conductance implies lower transmission probability and thus larger Fano factor (see Eq. 1). These results also indicate that current fluctuations persist with increasing wire length. For the one- and two-Si atom cases, the Fano factor is enhanced due to the fluctuations introduced by the proximity of the two electrodes. In these two cases, the distance between the electrodes is small so that the current across the electrodes without the Si atoms inbetween is of the same order of magnitude as the current of the total system (electrodes plus atoms). On the other hand, the shot noise of the bare electrodes without the atoms inbetween is larger than the shot noise of the total system. As a consequence, the Fano factor has a large contribution from the two bare electrodes, and the non-additive behavior of the total shot noise is evident in this case.

We now discuss the dependence of shot noise on the bias in the nonlinear regime. We choose a 3-Si wire as an example. The differential Fano factor (defined as $F = \partial S/\partial(2eI)$) as a function of bias is shown in Fig. 4. The differential conductance ($G = \partial I/\partial V$) is plotted in the same figure (right axis). From Fig. 4 it is evident that the differential conductance reaches a local minimum at about 2 V and a local maximum at about 3.5 V. The local minimum (maximum) in the conductance is associated to a lower (larger) density of states between the right and left Fermi levels (not shown). The differential Fano factor follows an opposite trend that can be explained again as
due to a decreased (increased) transmission probabilities for an electron scattering in the corresponding energy window. The Fano factor is also strongly enhanced in the regime where the differential conductance is close to zero. However, no complete suppression of noise is evident from Fig. 4 at the bias corresponding to the conductance peak. This is due to the contribution of several channels with different $K_\parallel$ that have a transmission probability lower than one at that bias.

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**FIG. 4:** Differential Fano factor (left axis) and differential conductance (right axis) as a function of applied bias in nonlinear regime for an atomic wire composed of three Si atoms placed between two electrodes as described in Fig. 1.