

# Full counting statistics of strongly non-Ohmic transport through single molecules

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We study analytically the full counting statistics of charge transport through single molecules, strongly coupled to a weakly damped vibrational mode. The specifics of transport in this regime – a hierarchical sequence of avalanches of transferred charges, interrupted by “quiet” periods – make the counting statistics strongly non-Gaussian. We support our findings for the counting statistics as well as for the frequency-dependent noise power by numerical simulations, finding excellent agreement.

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*Introduction.*—A prime qualitative difference of transport through single molecules as compared to artificial nanostructures lies in the role of the vibrational motion of the nuclei. This aspect is at the focus of current experiments [1, 2, 3], and is also being studied theoretically within a number of approaches [4, 5, 6, 7, 8, 9, 10]. The incorporation of molecular vibrations (phonons) into the theoretical description is mostly done within simplified (phenomenological) models, as opposed to purely electronic first-principles studies [11, 12].

Even within minimal models involving one molecular orbital coupled to a single vibrational mode, “unidirectional” transport (i.e., for voltages large compared to temperature) depends radically on the strength of the electron-phonon coupling, already at the qualitative level [6, 7, 10]. For weak and intermediate coupling [13], transport is adequately described in terms of individual electron transitions. By contrast when vibrational relaxation is slow, transport in the regime of strong electron-phonon coupling is appropriately captured within a scenario of *avalanches* of transferred electrons, with exponential spreads of height and duration [10].

More quantitatively, the time dependence of the current in the strong-coupling regime can be presented as

$$I(T) = f_1^{(0)}(T - t_1) + f_2^{(0)}(T - t_1 - t_2) + \dots, \quad (1)$$

where  $t_i$  are the time intervals between avalanches (quiet periods). These intervals are much longer than the typical duration  $\tau^{(0)}$  of an avalanche which occurs during the sparse periods when the vibrations are excited. The random function  $f_i^{(0)}(\tau)$  (which is nonzero only for times  $|\tau| \lesssim \tau^{(0)}$ ) describes the passage of a *large number*  $\int d\tau f_i^{(0)}(\tau) = N_i \gg 1$  of electrons through the molecule during the  $i$ th avalanche. Moreover, a numerical study of the avalanches [10] revealed their self-similar hierarchical structure, see Fig. 1. Quantitatively, this structure manifests itself in the fact that, during the time of avalanche  $\sim \tau^{(0)}$ , each function  $f_i^{(0)}$  itself takes the form of Eq. (1), with  $f_i^{(0)}$  replaced by random functions  $f_i^{(1)}$ , which describe avalanches of the *first generation* interrupted by quiet periods. Again, these quiet periods are much longer

than the characteristic time scale  $\tau^{(1)}$  of the functions  $f_i^{(1)}$ . For times shorter than  $\tau^{(1)}$ , the functions  $f_i^{(1)}$  have the form Eq. (1) with corresponding second-generation avalanches,  $f_i^{(2)}$ , having even shorter time-scale,  $\tau^{(2)}$ , and so on [14]. Numerical results supporting this scenario are shown in Fig. 1.

From the above discussion it is apparent that the statistical properties of charge transport through a molecule in the regime of strong electron-phonon coupling and through a conventional nanostructure are drastically different. For a nanostructure, all  $f_i^{(0)}(\tau)$  are  $\delta$  functions, so that  $N_i = 1$ . Hence, the distribution function  $P_T(Q)$  of the net transmitted charge  $Q$  during time  $T$  (full counting statistics [15]) is completely encoded in the statistics of the *waiting times*  $t_i$  for single-electron transitions between the leads. The distribution of waiting times reflects the details of the transport mechanism, and might be quite nontrivial [16]. Nevertheless, with all  $t_i$  being of the same order, the full counting statistics differs only weakly from a Gaussian distribution. These weak deviations are caused by correlations [17, 18], interactions [19], or the influence of the environment [20], and have been extensively studied theoretically.

By contrast, the counting statistics of avalanche-type transport is *insensitive* to the details of the passage of a single electron through the molecule, since the number of electrons involved in each avalanche is large. Instead, the counting statistics is governed *exclusively* by the transition rates between different vibrational states. These rates have a simple structure in the limit of strong coupling which allows us to develop a complete analytical theory for the regime of avalanche-type transport. In particular, in the present paper, we demonstrate that the full counting statistics  $P_T(Q)$  is given by a concise analytical expression, which is strongly skewed at “short” times ( $\sim$  zero-order quiet period) and evolves into a Gaussian only in the limit of very large  $T$ . Along with the counting statistics, we also study analytically how the hierarchy of avalanches manifests itself in the frequency dependence of the noise power  $S(\omega)$ . We support our analytical results with numerical Monte-Carlo (MC) simulations, finding

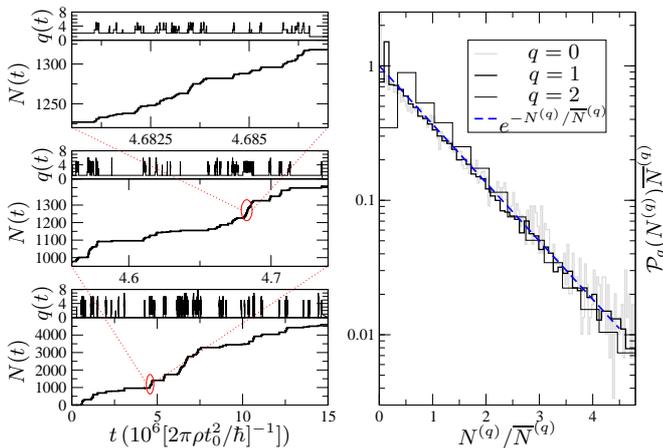


FIG. 1: Hierarchical character of transport. Left: Three generations of self-similar MC plots for  $\lambda = 4$  and  $eV = 3\hbar\omega_v$ , showing the net-transferred charge  $N$  and phonon state  $q$  as functions of time. ( $\omega_v$ : phonon frequency;  $\rho$ : density of states of leads;  $t_0$ : molecule-lead coupling). Right: Comparison of the fixed-point distribution for the transferred charge per generation- $q$  avalanche to numerical simulations for  $q = 0, 1, 2$  (mean values  $\overline{N}^{(0)} = 91.2$ ,  $\overline{N}^{(1)} = 11.1$ , and  $\overline{N}^{(2)} = 2.9$ ).

excellent agreement.

*Full counting statistics.*—Since different zeroth-generation avalanches are statistically independent, it is easy to derive a relation between the counting statistics  $P_T(Q)$  of the *net charge*  $Q$  and the conventional counting statistics  $\varphi_T(n)$  [15] of the *number* of zeroth-generation avalanches  $n$  during the time interval  $T$ . Indeed, using the definition  $P_T(Q) = \langle \delta(Q - \sum_{j=1}^n N_j) \rangle_{N_j, n}$ , and a Fourier representation of the RHS, one obtains

$$P_T(Q) = \int \frac{d\alpha}{2\pi} e^{i\alpha Q} \sum_n \left[ \tilde{\mathcal{P}}_0(\alpha) \right]^n \varphi_T(n), \quad (2)$$

where  $\tilde{\mathcal{P}}_0(\alpha) = \langle \exp(-i\alpha N_j) \rangle_{N_j}$  denotes the Fourier transform of the distribution function  $\mathcal{P}_0(N)$  of the total charge passing per zeroth-generation avalanche. The durations of the quiet periods obey Poisson statistics so that  $\varphi_T(n) = \exp(-\bar{n}_T) \bar{n}_T^n / n!$ . Here,  $\bar{n}_T$  denotes the average number of zeroth-generation avalanches within time  $T$ . Substituting this form into Eq. (2) and performing the summation over  $n$  yields an expression for the counting statistics similar to the Holtsmark distribution [21],

$$P_T(Q) = \int \frac{d\alpha}{2\pi} \exp \left\{ i\alpha Q + \bar{n}_T \left[ \tilde{\mathcal{P}}_0(\alpha) - 1 \right] \right\}. \quad (3)$$

Thus, the problem of the counting statistics reduces to finding the distribution function  $\mathcal{P}_0(N)$ . There are two facts that allow us to find this distribution, namely (i) the self-similar structure of avalanches and (ii) the large number  $n_q$  of generation- $(q+1)$  avalanches within a given generation- $q$  avalanche.

Our basic observation is that we can derive a recursion relation, relating the distribution functions  $\mathcal{P}_q(N)$

and  $\mathcal{P}_{q+1}(N)$  of the total passing charge ( $N^{(q)}$  and  $N^{(q+1)}$ , respectively) per avalanche for subsequent generations. This recursion follows from the obvious facts that  $N^{(q)} = \sum_{j=1}^{n_q} N_j^{(q+1)}$  and that different avalanches of a given generation are statistically independent. By analogy with the derivation of Eq. (2), we thus obtain

$$\mathcal{P}_q(N) = \int \frac{d\alpha}{2\pi} e^{i\alpha N} \sum_n \left[ \tilde{\mathcal{P}}_{q+1}(\alpha) \right]^n p_q(n), \quad (4)$$

where  $p_q(n)$  denotes the distribution function of  $n_q$ . To proceed further one has to specify the form of the distribution  $p_q(n)$ . This distribution is governed by the *microscopic* characteristics of the Franck-Condon transitions. We demonstrate below that  $p_q(n) = (1/\bar{n}_q) \exp(-n/\bar{n}_q)$ . Upon substituting this form into Eq. (4), the summation over  $n$  on the RHS can be easily performed and we obtain, after a Fourier transform of both sides,

$$\tilde{\mathcal{P}}_q(\alpha) = \frac{1}{\bar{n}_q} \frac{1}{1 - \tilde{\mathcal{P}}_{q+1}(\alpha) \exp(-1/\bar{n}_q)}. \quad (5)$$

The distribution  $\mathcal{P}_q$  can now be obtained from this equation by writing its general solution as  $\tilde{\mathcal{P}}_q(\alpha) = [1 + i\alpha\overline{N}^{(q)} + c_q(\alpha\overline{N}^{(q)})^2 + \dots]^{-1}$ . Inserting this into Eq. (5), we find that the numerical coefficients  $c_q$  flow to zero with  $q$  by virtue of the small parameter  $1/\bar{n}_q$ . Thus, the solution  $\tilde{\mathcal{P}}_q(\alpha) = [1 + i\alpha\overline{N}^{(q)}]^{-1}$  with Fourier transform  $\mathcal{P}_q(N) = \theta(N) \exp(-N/\overline{N}^{(q)})/\overline{N}^{(q)}$  can be viewed as a fixed point of the recursion equation Eq. (4) and since  $\overline{N}^{(q)} = \bar{n}_q \overline{N}^{(q+1)}$ , self-similarity is obeyed asymptotically. The existence of this fixed-point solution can be viewed as a consequence of remark (i) which implies that up to rescalings, the distribution functions  $\mathcal{P}_q(N)$  have the same functional form for *all*  $q$ . Fig. 1 numerically confirms this result for three different generations.

Once the form of  $\mathcal{P}_q(N)$  is established, the calculation of the counting statistics can be completed. Substituting  $\tilde{\mathcal{P}}_0(\alpha) = (1 + i\alpha\overline{N}^{(0)})^{-1}$  into Eq. (3), one can perform the integral and obtains

$$P_T(Q) = e^{-\bar{n}_T} \delta(Q) + e^{-\frac{\bar{n}_T}{\overline{N}^{(0)}} - \bar{n}_T} \sqrt{\frac{\bar{n}_T}{\overline{N}^{(0)}}} \frac{1}{Q} I_1 \left( 2\sqrt{\frac{\bar{n}_T Q}{\overline{N}^{(0)}}} \right). \quad (6)$$

Here  $I_1(z)$  denotes the modified Bessel function of order one. Eq. (6) is our central result. It is nicely confirmed by our MC results for the full counting statistics, as shown in Fig. 2, and describes the evolution of the counting statistics between two transparent limits:

(i) Short times,  $\bar{n}_T = T/t_i \ll 1$ : Using the expansion  $I_1(z) \approx z/2$  for  $z \ll 1$  we obtain from Eq. (6)

$$P_T(Q) \simeq e^{-\bar{n}_T} \left[ \delta(Q) + (\bar{n}_T/\overline{N}^{(0)}) \exp(-Q/\overline{N}^{(0)}) \right], \quad (7)$$

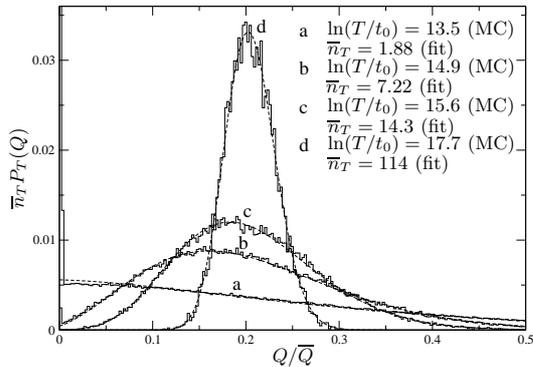


FIG. 2: Evolution of full counting statistics  $P_T(Q)$  for four time intervals  $T$  as indicated. MC data are fitted by Eq. (6).

Typically only a few electrons are transmitted through a molecule. The long tail described by the second term in Eq. (7) arises from realizations where an avalanche occurs within the time  $T$  and reflects the spread of charge within a single avalanche.

(i) Long times,  $\bar{n}_T \gg 1$ : Upon substituting the large- $z$  asymptote of  $I_1(z)$  into Eq. (6), it is easy to see that the second term has a sharp maximum centered at  $Q = \bar{n}_T \bar{N}^{(0)}$ , which is the average charge passed through the molecule after a large number of avalanches. Expansion of the exponent around the maximum yields the Gaussian distribution

$$P_T(Q) \simeq (\sqrt{\pi}\sigma_Q)^{-1} \exp\left[-\left(Q - \bar{n}_T \bar{N}^{(0)}\right)^2 / \sigma_Q^2\right] \quad (8)$$

with a width  $\sigma_Q = 2(\bar{n}_T)^{1/2} \bar{N}^{(0)}$ . This width is *twice* the width expected from the fluctuations of the waiting times. This enhanced broadening is due to fluctuations of the charge passed per avalanche. These additional fluctuations also manifest themselves in the noise characteristics of transport as analyzed below.

*Microscopic derivation of  $p_q(n)$ .*—The distribution

$p_q(n)$  is obtained by averaging the Poisson distribution of  $n$  for a *given* avalanche duration over the distribution of *durations*. On microscopic grounds, the latter distribution is simply exponential, which immediately implies  $p_q(n) = (1/\bar{n}_q) \exp(-n/\bar{n}_q)$  since  $\bar{n}_q$  is large.

To see this, we note that a generation- $q$  avalanche is interrupted when the molecule is deexcited from the maximal attained vibrational state  $q_{\max}$  back to state  $q$ . A molecule excited to  $q_{\max}$  has roughly similar rates for transitions to lower vibrational levels, while the rates for reexciting the molecule rapidly grow with  $q$ . Thus, to an excellent approximation the relative fractions of time spent in vibrational states higher than  $q$  during a generation- $q$  avalanche are governed by the excitation rates. In particular, the time spent in the vibrational level  $q_{\max}$  provides an effective waiting time for a transition back to phonon state  $q$ . Poisson statistics implies that this waiting time and hence the avalanche duration obey an exponential distribution.

*Noise spectrum  $S(\omega)$  of avalanche-type transport.*—We first derive a general expression for  $S(\omega)$  assuming arbitrary distributions  $\mathcal{P}_0(N)$  of the avalanche magnitudes and  $W(t)$  of the waiting times. For frequencies smaller than  $1/\tau^{(0)}$ , we have  $f_i^{(0)}(t) \simeq N_i \delta(t)$  in Eq. (1). Using Fourier representations of the  $\delta$  functions and averaging over the  $t_i$  and  $N_i$ , the average current becomes

$$\langle I(T) \rangle = \langle N_i \rangle \int \frac{d\alpha}{2\pi} e^{i\alpha T} \frac{\tilde{W}(\alpha)}{1 - \tilde{W}(\alpha)}, \quad (9)$$

where  $\tilde{W}(\alpha) = \langle \exp(-i\alpha t_i) \rangle_{t_i}$  denotes the Fourier transform of  $W(t)$ . In the long-time limit, only small values of  $\alpha$  contribute to the integral in Eq. (9) so that we can use the expansion  $\tilde{W}(\alpha) = 1 - i\alpha \langle t_i \rangle - (1/2)\alpha^2 \langle t_i^2 \rangle + \dots$ . Inserting this expansion into Eq. (9), keeping only the leading order in  $\alpha$  and performing the contour integration over  $\alpha$ , we recover the obvious result  $\langle I(T) \rangle = \langle N_i \rangle / \langle t_i \rangle$ .

Similarly, we express the current-current correlator as

$$\begin{aligned} \langle I(T_1) I(T_2) \rangle &= \int \frac{d\alpha}{2\pi} \frac{d\beta}{2\pi} e^{-i\alpha T_1 - i\beta T_2} \langle [N_1 e^{i\alpha t_1} + N_2 e^{i\alpha(t_1+t_2)} + \dots] [N_1 e^{i\beta t_1} + N_2 e^{i\beta(t_1+t_2)} + \dots] \rangle \\ &= \int \frac{d\alpha}{2\pi} \frac{d\beta}{2\pi} e^{-i\alpha T_1 - i\beta T_2} \left\{ \frac{\langle N_i \rangle^2 \tilde{W}(\alpha + \beta)}{1 - \tilde{W}(\alpha + \beta)} \left( \frac{1}{1 - \tilde{W}(\alpha)} + \frac{1}{1 - \tilde{W}(\beta)} - 1 \right) + \frac{(\langle N_i^2 \rangle - \langle N_i \rangle^2) \tilde{W}(\alpha + \beta)}{1 - \tilde{W}(\alpha + \beta)} \right\}. \quad (10) \end{aligned}$$

Here, the last equality follows upon term-by-term averaging and subsequent resummation of the series. To access the limit of long time  $T = (T_1 + T_2)/2$ , we introduce  $\omega = (\alpha - \beta)/2$  and  $\Omega = \alpha + \beta$ . Then, the exponent in the integrand in Eq. (10) assumes the form  $\exp(i\omega\tau - i\Omega T)$  with  $\tau = T_2 - T_1$ . The limit  $T \rightarrow \infty$  can now be taken

in a manner similar to the derivation of the average current above. Noting that the integrand can be directly identified with the noise spectrum  $S(\omega)$ , we find

$$S(\omega) = \frac{2}{\langle t_i \rangle} \left\{ \langle N_i \rangle^2 \left[ \frac{1}{1 - \tilde{W}(\omega)} + \frac{1}{1 - \tilde{W}(-\omega)} - 1 \right] \right.$$

$$+ (\langle N_i^2 \rangle - \langle N_i \rangle^2) \}. \quad (11)$$

Taking the zero-frequency limit requires one to keep terms of order  $\omega^2$  in the above-mentioned expansion of  $\tilde{W}(\pm\omega)$ . In this way, the Fano factor  $F = S(\omega = 0)/2e\langle I \rangle$  can be expressed as

$$F = \langle N_i \rangle \frac{\langle t_i^2 \rangle - \langle t_i \rangle^2}{\langle t_i \rangle^2} + \frac{\langle N_i^2 \rangle - \langle N_i \rangle^2}{\langle N_i \rangle}. \quad (12)$$

This equation allows for a transparent interpretation: Noise originates from two sources, namely the fluctuations in the intervals between avalanches and the fluctuations in the transmitted charge per avalanche. In the conventional situation where  $N_i = 1$  for all  $i$ , the Fano factor is given by the fluctuations of the waiting times  $t_i$  for a transition in which an electron passes either directly or sequentially from the left to the right lead. For example, for transport through a symmetric junction in the Coulomb-blockade regime, one immediately recovers  $F = 5/9$  [22] when taking into account that the rates of entering and leaving the dot are related as 2:1 due to spin. For the specific distributions adopted in our model, both terms in Eq. (12) contribute equally, and the Fano factor reduces to  $F = 2\overline{N}^{(0)}$ , which, in agreement with Eq. (8), is twice the value expected for a fixed magnitude of avalanches. This is confirmed by numerical results.

For frequencies larger than  $1/\tau^{(0)}$ , the ‘‘fine structure’’ of the avalanches described by the functions  $f_i^{(0)}$  in Eq. (1) must be taken into account. This fine structure can be incorporated into the noise spectrum Eq. (11) by replacing  $\langle N_i \rangle^2$  by  $\langle \tilde{f}(\alpha) \rangle \langle \tilde{f}(\beta) \rangle$  and  $\langle N_i^2 \rangle - \langle N_i \rangle^2$  by  $\langle \tilde{f}(\alpha) \tilde{f}(\beta) \rangle - \langle \tilde{f}(\alpha) \rangle \langle \tilde{f}(\beta) \rangle$ , where  $\tilde{f}(\alpha)$  denotes the Fourier transform. Explicitly employing the Poisson distribution of the waiting times leads to the remarkable simplification  $[1 - \tilde{W}(\omega)]^{-1} + [1 - \tilde{W}(-\omega)]^{-1} - 1 = 1$ . In this way, we obtain

$$S(\omega) = \frac{2}{\langle t_i \rangle} \langle \tilde{f}(\omega) \tilde{f}(-\omega) \rangle. \quad (13)$$

For frequencies of order  $\omega \simeq 1/\tau_0$  (where  $\tau_q$  denotes the average waiting time  $\langle t_i \rangle$  at level  $q$  of the hierarchy), we can ignore the fine structure of the avalanche and replace  $\tilde{f}(\omega) = N_i^{(0)}$ . Thus, we find  $S(\omega) = 2\langle [N_i^{(0)}]^2 \rangle / \tau_0$ . At higher frequencies  $\omega \simeq 1/\tau_1$ , the function  $f(t)$  is resolved into avalanches of generation  $q = 1$ . Then, we can write  $\langle \tilde{f}(\omega) \tilde{f}(-\omega) \rangle = \int dT \int d\tau e^{i\omega\tau} \langle f(T + \tau/2) f(T - \tau/2) \rangle$ . Up to the integral over  $T$ , this expression is analogous to  $S(\omega)$  itself, with zeroth-generation quantities replaced by corresponding  $q = 1$  quantities. For the frequencies of interest, we therefore find  $S(\omega) = (2/\tau_0)(\tau^{(0)} \langle [N_i^{(1)}]^2 \rangle / \tau_1)$ . Using the obvious relations  $\tau^{(0)} = \tau_1 \bar{n}_0$  and  $\overline{N}^{(0)} = \bar{n}_0 \overline{N}^{(1)}$  and generalizing to arbitrary  $q$ , we find

$$S_{q+1} = \frac{\overline{N}^{(q+1)}}{\overline{N}^{(q)}} S_q. \quad (14)$$

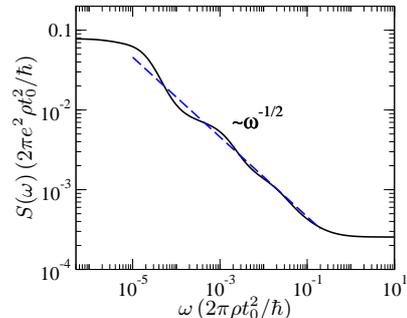


FIG. 3: Spectral density of noise in the Franck-Condon blockade regime for  $\lambda = 4$ ,  $eV = 3\hbar\omega_v$ . The observed approximate power law  $S(\omega) \sim \omega^{-1/2}$  arises from the hierarchical structure of avalanches.

Here, we define  $S_q = S(\omega \simeq 1/\tau_q)$  so that Eq. (14) provides a rule for extending the noise spectrum to progressively higher frequencies.

The essential *microscopic* inputs are the ratios  $\tau_{q+1}/\tau_q$  and  $\overline{N}^{(q+1)}/\overline{N}^{(q)}$ . Both ratios are determined by overlaps of displaced vibrational wavefunctions [10]. The rate  $1/\tau_q$  is dominated by the transition  $q \rightarrow q+1$ . Thus, it involves the overlap of *neighboring* harmonic oscillator states. By contrast,  $\overline{N}^{(q)}$  is inversely proportional to the transition rate from the maximal attained phonon levels to the  $q$ th vibrational level. The difference between these two rates is thus that the first involves four wavefunctions with index of order  $q$ , while the second involves only two. As a result, we can immediately establish from a quasiclassical evaluation of the matrix elements that  $\tau_q/(\overline{N}^{(q)})^2$  is essentially independent of  $q$ . With this input, we conclude that  $S(\omega) \sim \omega^{-\alpha}$  with exponent  $\alpha = 1/2$ . Since the noise power does not depend sensitively on  $\omega$  in finite intervals around  $1/\tau_q$ , this power law should be superimposed with steplike features in  $S(\omega)$ . As shown in Fig. 3, these conclusions agree with numerical simulations over several orders of magnitude in frequency.

*Conclusions.*—Our complete analytical description for the full-counting statistics and the frequency-dependent noise power of self-similar avalanche-type transport was made possible by the fact that current flow is essentially unidirectional. We emphasize that our arguments are quite general, with rather limited microscopic input, making our results potentially applicable far beyond the particular realization [10] of avalanche-type transport considered in the numerical simulations. Finally, we remark that direct vibrational relaxation (with rate  $\gamma$ ), neglected so far, only very gradually suppresses avalanche-type transport. Indeed, an immediate extension of our arguments for  $S(\omega)$  shows that the Fano factor decreases only as  $\sim 1/\gamma^{1/2}$ .

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- [1] H. Park, J. Park, A.K.L. Lim, E.H. Anderson, A.P. Alivisatos, and P.L. McEuen, *Nature* **407**, 57 (2000).
- [2] R.H.M. Smit, Y. Noat, C. Untied, N.D. Lang, M. van Hemert, J.M. van Ruitenbeek, *Nature* **419**, 906 (2002).
- [3] A.N. Pasupathy, J. Park, C. Chang, A.V. Soldatov, S. Lebedkin, R.C. Bialczak, J.E. Grose, L.A.K. Donev, J.P. Sethna, D.C. Ralph, and P.L. McEuen, *cond-mat/0311150* (2003).
- [4] L.I. Glazman and R.I. Shekhter, *Zh. Eksp. Teor. Fiz.* **94**, 292 (1987) [*Sov. Phys. JETP* **67**, 163 (1988)].
- [5] D. Boese and H. Schoeller, *Europhys. Lett.* **54**, 668 (2001).
- [6] S. Braig and K. Flensberg, *Phys. Rev. B* **70**, 085317 (2004).
- [7] A. Mitra, I. Aleiner, and A.J. Millis, *Phys. Rev.* **69**, 245302 (2004).
- [8] M. Galperin, M.A. Ratner, and A. Nitzan, *cond-mat/0405343* (2004).
- [9] S. Takei, Y.-B. Kim, and A. Mitra, *cond-mat/0412710*.
- [10] J. Koch and F. von Oppen, *cond-mat/0409667* (2004).
- [11] Y. Xue and M. Ratner, *cond-mat/0410539* (2004).
- [12] R.U.I. Liu, S.-H. Ki, H.U. Baranger, W. Yang, *cond-mat/0409523* (2004).
- [13] Weak (strong) coupling corresponds to  $\lambda \ll 1$  ( $\lambda \gg 1$ ), where the dimensionless electron-phonon coupling  $\lambda$  equals the shift of the equilibrium nuclear normal-mode variable between neutral and charged molecule, measured in units of the spatial extent of the vibrational ground-state wavefunction.
- [14] The hierarchy breaks off once the transferred charge per avalanche is of order 1.
- [15] L.S. Levitov and G.B. Lesovik, *JETP Lett.* **58**, 230 (1993).
- [16] Ya.M. Blanter and M. Büttiker, *Phys. Rep.* **336**, 1 (2000).
- [17] L.S. Levitov and M. Reznikov, *Phys. Rev. B* **70**, 115305 (2004), and references therein.
- [18] U. Gavish, Y. Imry, Y. Levinson, and B. Yurke, in *Quantum Noise in Mesoscopic Physics*, ed. by Yu.V. Nazarov (Kluwer, Dordrecht, 2003).
- [19] M. Kindermann and Yu.V. Nazarov *Phys. Rev. Lett.* **91**, 13682 (2003).
- [20] M. Kindermann, Yu.V. Nazarov, and C.W.J. Beenakker, *Phys. Rev. B* **69**, 035336 (2004).
- [21] J. Holtsmark, *Ann. Physik* **58**, 577 (1919).
- [22] Yu.V. Nazarov and J.J.R. Struben, *Phys. Rev. B* **53**, 15466 (1996).