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# Indications of Universal Excess Fluctuations in Nonequilibrium Systems

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The fluctuation in electric current in nonequilibrium steady states is investigated by molecular dynamics simulation of macroscopically uniform conductors. At low frequencies, appropriate decomposition of the spectral intensity of current into thermal and excess fluctuations provides a simple picture of excess fluctuations behaving as shot noise. This indicates that the fluctuation–dissipation relation may be violated in a universal manner by the appearance of shot noise for a wide range of systems with particle or momentum transport.

KEYWORDS: nonequilibrium steady state, fluctuation–dissipation relation, shot noise, electric conduction
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In equilibrium states, the fluctuation of an observable is related *universally* to a linear response function by the fluctuation–dissipation relation (FDR).[1] In nonequilibrium steady states (NESSs), the FDR is often violated, and 'excess fluctuation' (XF) appears.[2–17] XF plays crucial roles in many fields of physics, including single electron tunneling,[2] the squeezing of photons,[3,4] the measurement of fractional charge,[5] and the determination of the fundamental limits of quantum interference devices.[6] However, unlike equilibrium fluctuation, it is not yet well understood whether *universal* properties exist in XF.[18]

Experimentally, the FDR violation is hardly observable in heat conduction because a convection current or a phase transition is induced for large temperature difference (which drives heat conduction) before XF becomes detectable. In contrast, the violation has been widely observed in *systems with particle (or momentum) transport*, such as electric conductors and photoemitting devices.[3–17] We therefore consider such systems.

Among such systems are simple systems, including mesoscopic conductors,[5–15] conductors with junctions[11,16,17] (e.g., tunnel and PN junctions), and light-emitting diodes.[3,4] These systems are simple in the sense that the number of electron modes is small and/or many-body interactions are unimportant and/or dissipation is negligible and/or the principal origin of XF is localized in certain mesoscopic regions. XF generated in such a case takes the form of shot noise.[3–17] Here, the term 'shot noise' is used in a wide sense, which stands for fluctuation whose spectral intensity $S_I$ is proportional to the absolute value of average flux, $|\langle I\rangle|$.[19] The ratio $W$ of $S_I$ to its Poissonian value is called the Fano factor, which takes various values depending on the details of the systems.[3–17]

The situation is completely different for *uniform macroscopic* conductors, for which the assumptions made in refs. 3–17 do not hold. Although the FDR violation is hardly observable in uniform metals, it is widely observed in uniform semiconductors.[16] Most experiments on the latter showed that XF is dominated by $1/f$ noise, which is proportional to $\langle I\rangle^2$.[16] Although shot noise may also exist in such systems, it would be masked by $1/f$ noise[20] because the latter increases more rapidly with increasing $|\langle I\rangle|$. However, the origin of $1/f$ noise is believed to be imperfections in samples, such as the fluctuation in carrier number and the migration of impurities, which result in a strong sample dependence of $1/f$ noise.[16] Since imperfections in samples are of secondary interest in fundamental physics (nonequilibrium statistical mechanics), a natural question arises: What fluctuation appears in perfect samples? In this paper, we address this question and report a property of XF that may be universal.

The models and results of the previous works on mesoscopic conductors[5–15] are not applicable to macroscopic conductors, because, as mentioned above, many assumptions that do not hold in macroscopic conductors have been made in those works. We therefore take a different approach. That is, we use molecular dynamics (MD) simulation on a model that we believe captures the essential elements of macroscopic conductors.[21,22] This enables us to study the NESSs of perfect samples, without making the assumptions made in the works on mesoscopic conductors. Since we can vary the values of the parameters to a great extent, we are able to present results that may indicate a universal character.

Except at low temperatures, quantum effects seem to play minor roles in macroscopic conductors far from equilibrium, because of the strong decoherence. We therefore use the classical model of electric conduction proposed in ref. 21, which describes doped semiconductors at room temperature well.[22] The system includes three types of classical particles, which we call electrons (each with mass $m_{\mathrm{e}}$ and charge $e$), phonons (each with mass $m_{\mathrm{p}}$), and impurities. Their number densities are denoted by $n_{\mathrm{e}}$, $n_{\mathrm{p}}$, and $n_{\mathrm{i}}$, respectively. For simplicity, we assume a two-dimensional system, the size of which is $L_x \times L_y$. In the $x$-direction, we apply an external electric field acting *only on electrons*, and impose the periodic boundary condition. The boundaries in the $y$-direction are potential walls for electrons and thermal walls for phonons. The thermal walls reflect phonons with random velocities sampled from an equilibrium distribution with temperature $T_0$.[21] This enables phonons to carry heat constantly out of the system thereby keeping the system in a NESS. Impurities are immobile and play a role of providing random potential.

We assume short-range interactions among *all* particles. Since interaction potential is well characterized by its scattering cross section, detailed forms of the potential are

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expected to be irrelevant when studying general nonequilibrium properties. Therefore, we here take a simple form, $k_0(\max\{0, d_{jl}\})^{5/2}$. Here, $k_0$ is a constant and $d_{jl} = R_j + R_l - |\boldsymbol{r}_j - \boldsymbol{r}_l|$ is the overlap of the potential ranges. $R_j$ is the radius of the potential range ($R_\mathrm{e}$, $R_\mathrm{p}$, and $R_\mathrm{i}$ for an electron, phonon, and impurity, respectively), and $\boldsymbol{r}_j$ is the position of the $j$-th particle. We can change the strength of scattering by varying $R_j$ (and particle number density).

This model corresponds to a perfect sample because the total number of carriers does not change and because impurities do not move. This system is macroscopically uniform although the translational invariance is broken by impurities and the thermal walls for phonons. Furthermore, the model and results are also applicable to systems that have a mass flow of neutral particles.[22)]

We use units in which $m_\mathrm{e}$, $R_\mathrm{e}$, $e$, the Boltzmann constant, and a reference energy are unity. Regarding the other parameters, the main result, eq. (5), is insensitive to their values, as will be shown later. We here fix $R_\mathrm{p} = 1$, $T_0 = 1$, and $k_0 = 4000$; the other parameters are varied to illustrate the possible universality of the result.

To investigate nonequilibrium states of this model, we perform MD simulation using Gear's fifth-order predictor–corrector method.[21)] The time-step width is set to $10^{-3}$. The initial position of each particle is randomly arranged so as not to be in contact with the other particles, and the initial velocities of the electrons and phonons are given by the Maxwell distribution with temperature $T_0$. We calculate various quantities after the system reaches a NESS.

The electric field applied to the system is composed of a time-independent field $E$, which is varied in a wide range, and a time-dependent field $\varepsilon f(t)$, which is small. The electric field induces electric current $I(t) \equiv e n_\mathrm{e} L_y V_\mathrm{e}^x(t)$, where $V_\mathrm{e}^x$ is the velocity in the $x$-direction (i.e., along the electric field) of the center of mass of electrons. We take $\varepsilon \neq 0$ only when we calculate the *differential* response function $\mu(t-\tau; E)$ *of a NESS*, which is defined by

$$\langle \delta I(t) \rangle_{E,\varepsilon} = \int_{-\infty}^{t} \mathrm{d}\tau\, \mu(t-\tau; E) L_x \varepsilon f(\tau) + O(\varepsilon^2), \qquad (1)$$

for $t > \tau$ and by $\mu(t-\tau; E) = 0$ for $t < \tau$. Here, $\delta I = I - \langle I \rangle_{E,0}$, and $\langle \cdots \rangle_{E,\varepsilon}$ denotes the average at the NESS in the electric field $E + \varepsilon f(t)$. The convolution theorem yields $\tilde{\mu}(\omega; E) = \lim_{\varepsilon\to 0} \langle \delta \tilde{I}(\omega) \rangle_{E,\varepsilon} / L_x \varepsilon \tilde{f}(\omega)$, where the tilde denotes the Fourier transform. Note that $\tilde{\mu}(\omega; E)$ differs from that in an *equilibrium state*, $\tilde{\mu}(\omega; 0)$.

We are mainly interested in the current fluctuation that is characterized by the spectral intensity $S_I(\omega; E)$ of $I(t)$ for $\varepsilon = 0$. By the Wiener–Khinchine theorem,[1)] $S_I(\omega; E)$ is equal to the Fourier transform of the autocorrelation function $\langle \delta I(t) \delta I(0) \rangle_{E,0}$ of current. In equilibrium states ($E = 0$), the FDR, $S_I(\omega; 0) = 2T\,\mathrm{Re}\,\tilde{\mu}(\omega; 0)$, holds for all $\omega$.[1)] Here, $T$ is the temperature of the conductor, which is equal to $T_0$ when $E = 0$. We plot both sides of this relation in Fig. 1(a), and confirm that it holds in our simulation.

When larger $E$ ($\neq 0$) is applied, $\langle I \rangle_{E,0}$ becomes nonlinear with $E$, as shown in the inset of Fig. 1(a). In such NESSs, we find that the FDR is violated, i.e., for any $T$ that is independent of $\omega$,

$$S_I(\omega; E) \neq 2T\,\mathrm{Re}\,\tilde{\mu}(\omega; E) \text{ for some } \omega. \qquad (2)$$

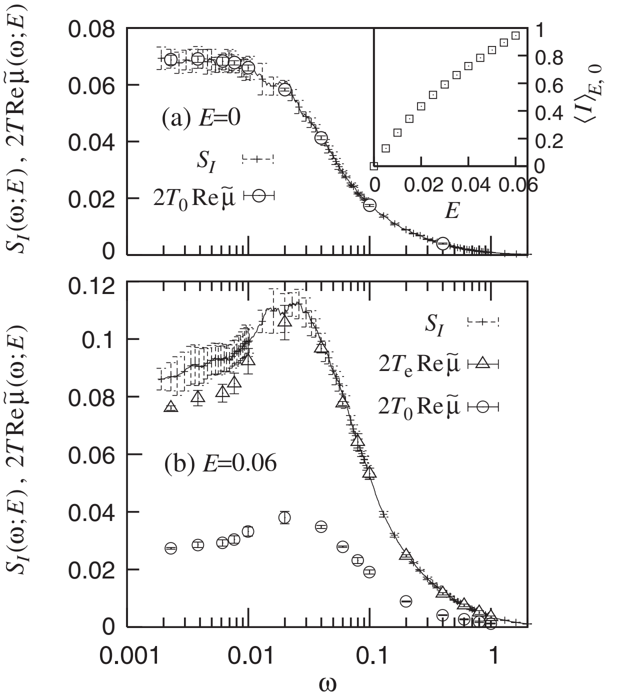


Fig. 1. (a) $S_I(\omega; E)$ and $2T_0\,\mathrm{Re}\,\tilde{\mu}(\omega; E)$ for $E = 0$. The inset shows $\langle I \rangle_{E,0}$ versus $E$. (b) $S_I(\omega; E)$, $2T_\mathrm{e}(E)\,\mathrm{Re}\,\tilde{\mu}(\omega; E)$, and $2T_0\,\mathrm{Re}\,\tilde{\mu}(\omega; E)$ for $E = 0.06$ (nonlinear response regime). In these simulations, $m_\mathrm{p} = 1$, $R_\mathrm{i} = 0.5$, $L_x = 750$, $L_y = 125$, $n_\mathrm{e} = n_\mathrm{p} = 0.016$, and $n_\mathrm{i} = 2/375$. The data points are the averages of five samples (impurity configurations) and the error bars are the standard deviations among them.

This is demonstrated in Fig. 1(b), which shows $S_I(\omega; E)$, $2T_0\,\mathrm{Re}\,\tilde{\mu}(\omega; E)$, and $2T_\mathrm{e}(E)\,\mathrm{Re}\,\tilde{\mu}(\omega; E)$ in a nonlinear response regime. Here, $T_\mathrm{e}(E) \equiv m_\mathrm{e} \langle (v_\mathrm{e}^x - \langle v_\mathrm{e}^x \rangle_{E,0})^2 \rangle_{E,0}$ is a kinetic temperature of electrons ($v_\mathrm{e}^x$ is the velocity of an electron in the $x$-direction). When we employ $2T_0\,\mathrm{Re}\,\tilde{\mu}(\omega; E)$ as the right-hand side (RHS) of the FDR, the violation of the FDR is observed in a wide frequency range. When we use $2T_\mathrm{e}(E)\,\mathrm{Re}\,\tilde{\mu}(\omega; E)$ as the RHS, the violation is observed at low frequencies ($\omega \ll \omega_0$)[23)] while the RHS coincides with $S_I(\omega; E)$ at higher frequencies ($\omega \gg \omega_0$), where $\omega_0$ is the crossover frequency between the regimes of FDR violation and validation. These data also show that the FDR is violated for *any definitions of* $T$ that is independent of $\omega$.

Now we discuss the main finding of this paper. Since we have seen that the FDR violation is manifested at lower frequencies, we look at the low-frequency region ($\omega \ll \omega_0$). Among many possible definitions of 'thermal fluctuation' of $I$ for $E \neq 0$, we employ

$$S_\mathrm{th}(\omega; E) \equiv 2T_0\,\mathrm{Re}\,\tilde{\mu}(\omega; E), \qquad (3)$$

which is the RHS of eq. (2) with $T = T_0$. Using this, we decompose the total fluctuation $S_I$ into two parts:

$$S_I(\omega; E) = S_\mathrm{th}(\omega; E) + S_\mathrm{exs}(\omega; E). \qquad (4)$$

Since the thus-defined $S_\mathrm{exs}$ quantifies the FDR violation, we call it excess fluctuation. In Fig. 2, we plot $S_\mathrm{exs}$ for $\omega \simeq 0.002$ *as a function of* $\langle I \rangle_{E,0}$. [We can translate a function of $E$ into a function of $\langle I \rangle_{E,0}$ because of the one-to-one correspondence between $E$ and $\langle I \rangle_{E,0}$.] Since the FDR holds in equilibrium states, $S_\mathrm{exs} \simeq 0$ when $\langle I \rangle_{E,0}$ is small. As $\langle I \rangle_{E,0}$ increases, $S_\mathrm{exs}$ exhibits a crossover behavior from near equilibrium to far from equilibrium as

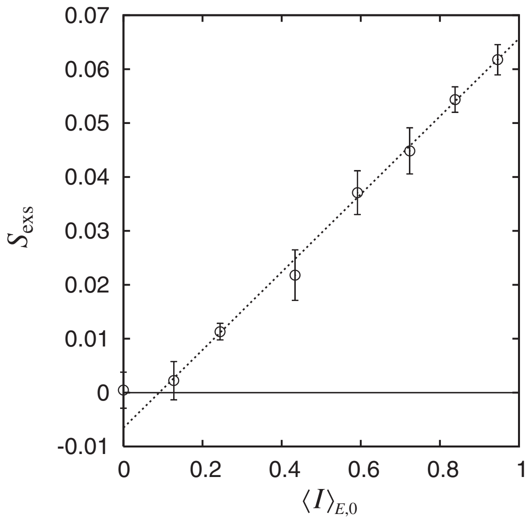


Fig. 2. Excess fluctuation $S_{exs}$ at a low frequency, plotted against $\langle I\rangle_{E,0}$. The dotted line represents the asymptote, $W(|\langle I\rangle_{E,0}| - I_0)$, fitted with the four data points at larger values of $\langle I\rangle_{E,0}$. The parameters of the simulation and the meaning of the error bars are the same as those in Fig. 1.

$$S_{exs} \simeq \begin{cases} 0 & (|\langle I\rangle_{E,0}| \ll I_0), \\ W(|\langle I\rangle_{E,0}| - I_0) & (|\langle I\rangle_{E,0}| \gg I_0), \end{cases} \tag{5}$$

where $I_0$ is a certain crossover value of the current. In the latter region ($|\langle I\rangle_{E,0}| \gg I_0$), $S_{exs}$ takes the form of shot noise, where $W$ is the Fano factor.[3–11,16,17)]

We have thus found that the dominant mechanism that breaks the FDR is the appearance of shot noise. To confirm that this observation holds widely for the model considered here, we also study $S_{exs}$ in the following cases: (i) another impurity density, $n_i = 0.016$, (ii) other linear dimensions $L_x$ (along $E$) of the system, $L_x = 375$, 300, 187.5, and 150, and (iii) the values of the other parameters are changed significantly (e.g., $n_e = 0.008$, $m_p = 10$, and $R_i = 2$). (iv) The thermal walls for phonons are set away from the boundaries for electrons, as shown in the top-left inset of Fig. 3(b).

Figure 3 shows the results in case (iv). In this case, the local phonon temperature $T_p$ around the boundaries for electrons is markedly different from $T_0$, as shown in Fig. 3(a), where $T_p \equiv m_p\langle(v_p^x - \langle v_p^x\rangle_{E,0})^2\rangle_{E,0}$ ($v_p^x$ is the local phonon velocity in the $x$-direction). Despite this fact, $S_{exs}$ is well-fitted again by eq. (5), as shown in Fig. 3(b), if we define thermal fluctuation again by eq. (3) using $T_0$. Furthermore, we have found, although the data are not shown here, that eq. (5) also holds well in cases (i)–(iii) (except when the densities are so high that a liquid-solid phase transition takes place). Note in particular that the validity of eq. (5) in case (iii) suggests that it holds independently of details of the models, because case (iii) naturally includes, for example, the case where the $R_j$ are specific functions of $n_e$.

The above observations strongly indicate the robustness of eq. (5). Note that this possible universality is visible only when thermal fluctuation in nonequilibrium states is appropriately defined as eq. (3). In fact, we have found (although the data are not shown here) that the possible universality is obscured if we use $T_e(E)$ instead of $T_0$ in thermal fluctuation.

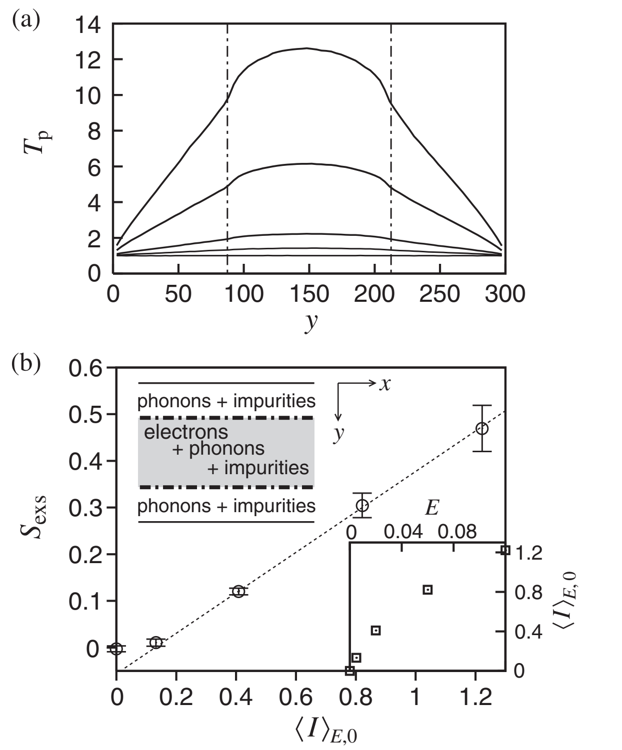


Fig. 3. (a) Local phonon temperature $T_p$ and (b) excess fluctuation $S_{exs}$ for $\omega \simeq 0.002$, plotted against $\langle I\rangle_{E,0}$, for a system where the thermal walls (at $y = 0$ and 300) for phonons are set away from the potential walls (at $y = 87.5$ and 212.5, dash-dotted lines) for electrons, as shown in the top-left inset of (b). In (a), the solid lines from bottom to top correspond to the data for $E = 0$, 0.01, 0.02, 0.06, and 0.12. In (b), the dotted line represents the asymptote $W(|\langle I\rangle_{E,0}| - I_0)$. The meaning of the error bars is the same as that of the error bars in Fig. 1. Bottom-right inset: $\langle I\rangle_{E,0}$ versus $E$ for this system. We take $m_p = 1$, $R_i = 0.5$, $L_x = 375$, $n_e = 0.016$, $n_p = 19/1125$, and $n_i = 2/375$.

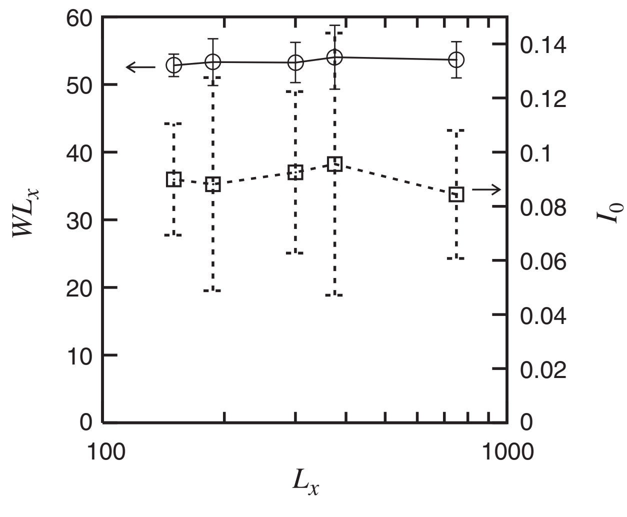


Fig. 4. $WL_x$ (circles; left axis) and $I_0$ (squares; right axis) for several sizes of the system. The particle densities are the same as those in Fig. 1.

Using the results in case (ii), we also investigate the $L_x$ dependences of $W$ and $I_0$. We evaluate $W$ and $I_0$ by fitting the numerical results of $S_{exs}$ for large $|\langle I\rangle_{E,0}|$ to the asymptotic form of eq. (5). In Fig. 4, we show $WL_x$ and $I_0$ versus $L_x$. We see that $WL_x$ is almost independent of $L_x$, i.e., $W \sim 1/L_x$. This agrees with the partial result for macroscopic conductors in ref. 9 (however, see note[24]), and coincides with the results for long mesoscopic conductors.[7,8] Furthermore, we observe that $I_0$ is almost independent of $L_x$, although the error bars are somewhat large.

By combining the present results with the results on simple systems,[3–17] we conjecture that the FDR is violated not in a random and system-dependent manner but in a universal manner by the appearance of shot noise, for a wide range of systems from mesoscopic to macroscopic. All details of individual systems are absorbed into $W$, $I_0$, and the differential response function $\mathrm{Re}\,\tilde{\mu}$ (by which thermal fluctuation is defined). The origin of current fluctuation in the present model is the chaotic behavior of interacting many particles in classical systems, while that in the simple systems[3–17] is essentially the probabilistic nature of quantum or thermal-activation processes of noninteracting particles. Despite such a marked difference, $S_{\mathrm{exs}}$ takes an identical form in all systems. This observation may be used as a touchstone in nonequilibrium thermodynamics or statistical mechanics beyond the linear response theory.

Note that the present results could never be obtained by a naïve perturbation expansion, in powers of the driving force $E$, about an equilibrium state. For example, the relation $S_{\mathrm{exs}} \propto |\langle I \rangle|$ suggests that such a power series would not converge for large $E$. Using MD simulation, we have successfully investigated such a "non-perturbative regime". Our results may be confirmed experimentally, for example, in high-quality doped semiconductors, which may be prepared by modulation doping, at room temperature.

In conclusion, we have presented a study of excess fluctuations in a nonequilibrium system and found that the fluctuation–dissipation relation is violated in a manner that may be universal. We hope that our work will stimulate further research that will test the correctness of this conjecture for wider classes of systems.

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18) Although the fluctuation theorem [see, for example, D. J. Evans and D. J. Searles: Adv. Phys. **51** (2002) 1529] would formally hold in NESSs, it cannot be used to treat or predict XF discussed in refs. 2–17 and here.
19) In some mesoscopic conductors, transmittance $\mathcal{T}$ varies as a function of applied voltage, and so does $W$ $(= 1 - \mathcal{T})$. Although $S_I$ then becomes a nonlinear function of $|\langle I \rangle|$, it is also called shot noise because the physics involved is the same as that in the other cases where $\mathcal{T}$ is constant or small.
20) Although $1/f$ noise also appears in mesoscopic conductors, it is sufficiently small in good samples for shot noise to be observed.[5,7–11]
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22) T. Yuge and A. Shimizu: Prog. Theor. Phys. Suppl. **178** (2009) 64. Note that Umklapp scattering is unimportant in electron transport in semiconductors because their average wave number is much smaller than the sizes of reciprocal lattice vectors.
23) In the two-sided Welch's test, the null hypothesis that $S_I(\omega \simeq 0; E = 0.06) = 2T_{\mathrm{e}}\,\mathrm{Re}\,\tilde{\mu}(\omega \simeq 0; E = 0.06)$ is rejected at the 99.9% confidence level.
24) The derivation of the scaling law in refs. 9 and 11 is rigorous in the macroscopic regime *if shot noise is dominant*. However, similarly to other works, refs. 9 and 11 did not show that shot noise is indeed dominant in macroscopic conductors.