Photovoltaic and Rectification Currents in Quantum Dots

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(Dated: October 1, 2004)

We investigate theoretically and experimentally the statistical properties of dc current through an open quantum dot subject to ac excitation of a shape-defining gate. The symmetries of rectification current and photovoltaic current with respect to applied magnetic field are examined. Theory and experiment are found to be in good agreement throughout a broad range of frequency and ac power, ranging from adiabatic to nonadiabatic regimes.

Transport in mesoscopic systems subject to time-varying fields combines elements of non-equilibrium physics and quantum chaos. This combination extends the scope of mesoscopic physics and is likely to be important in quantum information processing, where fast gating and quantum coherence are both required. Of particular importance is the ability to control external fields applied to the mesoscopic system and to distinguish effects of these fields on quantum dynamics of the system. For example, two distinct contributions to direct current through an open quantum dot due to an oscillating perturbation have been identified1,2 and observed experimentally in Ref. 3.

In this Letter, we investigate the statistical properties of dc currents resulting from an applied ac electric field over a wide range of excitation frequencies, paying particular attention to the presence or absence of symmetry with respect to magnetic field in various regimes. Theoretical analysis is based on recently developed time-dependent random matrix theory4,5. Experiments are performed with a gate-defined GaAs quantum dot subject to ac excitation of a gate at MHz to GHz frequencies. At low excitation frequencies $\omega \ll \tau_d^{-1}$ ($\tau_d$ is the electron dwell time in the dot) the present theoretical results are consistent with those obtained by adiabatic approximations1,3. However, the analysis is applicable over a wider range of frequencies $\omega \lesssim E_T$, where $E_T = \hbar / \tau_{cross}$ is the Thouless energy and $\tau_{cross}$ is the electron crossing time of the dot. At higher frequencies $\omega \gtrsim E_T$, the system may be studied by methods developed for bulk conductors6,8.

Three distinct contributions to dc current through the dot can be identified, resulting from: i) an applied dc bias; ii) an ac bias at the excitation frequency (i.e., rectification effects6,7); iii) photovoltaic effects5,8. We restrict our attention to one-parameter excitation, noting that while in the adiabatic regime one- and two-parameter excitations affect the system differently, beyond the adiabatic regime, $\omega \gtrsim \tau_d^{-1}$, the differences disappear5.

The Hamiltonian of electrons in the dot in the presence of a magnetic flux $\Phi$ is represented by a Hermitian $M \times M$ matrix $\hat{H}(t) = \hat{H}_\Phi + \hat{V} \cos \omega t$, with the time independent part $\hat{H}_\Phi$ being a random realization of a matrix from a Gaussian unitary ensemble with the mean level spacing $\delta_1$, and $\hat{V}$ being a matrix from a Gaussian orthogonal ensemble characterized by the strength $C_0 = \pi \text{Tr} \hat{V}^2 / M^2 \delta_1$ and $M \delta_1 \sim E_T$. The parameter $C_0$ determines the energy displacement of an electron state due to the applied perturbation $\hat{V}$. The contact between the left (right) lead and the dot contains $N_l$ ($N_r$) open channels, we enumerate channels, $\alpha$, in the left ($\alpha = 1 \ldots N_l$) and the right ($\alpha = N_l + 1 \ldots N_{ch}$) contacts, $N_{ch} = N_l + N_r$. The corresponding experimental setup is shown in Fig. 1.

The dc current $I^\Phi$ through the dot is determined by the scattering matrix [$S_{\Phi}(t, t')$]$_{\alpha\beta}$, see Ref. 5:

$$I^\Phi = \frac{e \omega}{2\pi} \int_0^{2\pi/\omega} dt \int_{-\infty}^{+\infty} dt_1 dt_2 \times \text{Tr} \left\{ \hat{f}(t_1, t_2) \left[ S_\Phi(t_2, t) \Lambda S_\Phi(t, t_1) - \Lambda \delta_{t_1 t_2} \delta_{t, t_2} \right] \right\}. \quad (1)$$

Here $\delta_{t_1 t_2} = \delta(t - t')$ and

$$f_{\alpha\beta}(t_1, t_2) = \frac{k_B T}{\hbar} \Lambda_{\alpha\beta} \exp \left( \frac{i}{\hbar} \int_{t_2}^{t_1} V_\alpha(\tau) d\tau \right) \frac{\sinh(\pi k_B T(t - t')/\hbar)}{\sinh(\pi k_B T(t - t')/\hbar)} \quad (2)$$

is the distribution function of electrons in channel $\alpha$ at temperature $T$ and voltage $V_\alpha(t)$. At sufficiently low frequencies $\omega \ll E_c/\hbar$ ($E_c$ is the dot charging energy) $V_\alpha(t)$ is simply related to the bias $V(t)$ across the dot: $V_\alpha(t) = \Lambda_{\alpha\alpha} V(t)$. Elements of the diagonal matrix $\Lambda$ are $\Lambda_{\alpha\alpha} = N_r / N_{ch}$ for $1 \leq \alpha \leq N_l$, and $\Lambda_{\alpha\alpha} = -N_l / N_{ch}$ for $N_l < \alpha \leq N_{ch}$.

We consider the bias $V(t)$ across the dot in the form $V(t) = V_0 + V_\omega \cos(\omega t + \varphi_1)$. The dc current through the dot to first order in dc bias $V_0$ and ac bias $V_\omega$ is 11

$$I^\Phi = I_{\text{ph}}^\Phi + I_{\text{1}}^\Phi + g_0^\Phi V_0, \quad I_{\text{1}}^\Phi = g_1^\Phi V_\omega, \quad (3)$$

where the first term represents the photovoltaic current $I_{\text{ph}}^\Phi = I^\Phi (V_\omega \equiv 0)$, see Eqs. (1) and (2). The second and third terms in Eq. (3) represent the contributions to the
current due to dc bias $V_0$ and ac bias $V_\omega$, respectively:
\[
\delta G_0^\Phi = \frac{e^2}{\pi h} \left[ G_0 - \delta G_0^\Phi(t) \right], \quad \delta G_1^\Phi = -\frac{e^2}{\pi h} \delta G_1^\Phi(t),
\]
where $G_0 = N_l N_r / N_{ch}$ is the classical conductance and $\delta G_k(t) = \int_0^{2\pi / \omega} \delta G_k(t) \omega dt / 2\pi \pi$ stands for time averaging of the “instantaneous conductance” ($k = 0, 1$)
\[
\delta G_k^\Phi(t) = \int d\tau F_k(\tau) \int d\theta \cos(k \omega \theta + \varphi_k))
\times \text{Tr} \left\{ \hat{\Delta}^\Phi_\omega \left( \theta - \frac{T}{2}, t \right) \hat{\Delta}^\Phi_\omega \left( t, \theta + \frac{T}{2} \right) \right\},
\]
\[
F_0(\tau) = \frac{\pi k_B T \omega}{\sinh(\pi k_B T \tau / \hbar)}, \quad F_1(\tau) = \frac{2 \pi k_B T \sin(\omega \tau / 2)}{\hbar \omega \sinh(\pi k_B T \tau / \hbar)}.
\]
We observe that $\delta G_1^\Phi(t) = \delta G_0(t) \cos(\omega t + \varphi_1)$ in the adiabatic limit $\hbar \omega \ll \max\{N_{ch} \delta_1, k_B T\}$ considered in Refs. [2, 12].

Below we study the variance of the photovoltaic current $\langle I_1^\Phi \rangle$ and the conductances $\delta G_0^\Phi$ and $\delta G_1^\Phi$ with respect to random realizations of the Hamiltonian $\hat{H}_\Phi$. Following Refs. [2, 12], we find in the limit $N_{ch} \gg 1$ and at magnetic fields $\Phi \gg \Phi_0 \sqrt{N_{ch} / M}$ destroying the weak localization ($\Phi_0 = h c / e$)
\[
\langle (\hat{I}_1^\Phi)^2 \rangle = \frac{G_0 e^2 \omega^4 C_0 \delta_1}{2 \pi^3 h^2} \int \frac{d\omega dt}{4 \pi^2} \int \frac{d\theta}{\pi} \int \frac{d\theta'}{\pi} K \left[ B_{t'\tau}^\Phi - B_{t\tau}^\Phi \right],
\]
and $\langle \hat{I}_1^\Phi \hat{I}_1^{-\Phi} \rangle = 0$, see Ref. [4]. In Eqs. [4] and [7] the angle brackets $(\ldots)$ stand for the averaging with respect to realizations of $\hat{H}_\Phi$. Functions $B_{t'\tau}^\Phi$ and $B_{t\tau}^\Phi$ describe the distribution function [14] of electrons in the dot in the presence of time-dependent electric fields and kernels $K^\pm \equiv K_{t'\tau, \tau}^\pm$ describe the evolution of electron states [15] in these fields. Both functions $B_{t'\tau}^\Phi$ and $B_{t\tau}^\Phi$ and the kernels $K_{t'\tau, \tau}^\pm$ contain the diffusion $D(t_1, t_2, \tau) = \exp \left\{ -\int_{t_1}^{t_2} \Gamma(\tau, t) dt \right\}$ or the Cooperon $C(t_1, t_2, \tau) = \exp \left\{ -\frac{1}{2} \int_{t_1}^{t_2} \Gamma(\tau, t) dt \right\}$.

In the experiment, the ac bias $V_\omega$ results from capacitive coupling between the leads and the gate on which the ac voltage is applied (see the inset in Fig. 1). Therefore $V_\omega$ is proportional to the amplitude of the ac voltage at the gate. Assuming that $C_0$ is linear in the applied power to the gates, we write $V_\omega = C_0 / \gamma_0$. The coefficient $\gamma_0$ has units of voltage and is independent of realizations of the quantum dot (we disregard fluctuations of $V$ over different realizations of $\hat{H}_\Phi$). Therefore, the correlation functions of the rectification current $I_1^\Phi = \delta G_1^\Phi V_\omega$ are determined by the correlators of $\delta G_1^\Phi$:
\[
\langle \hat{I}_1^\Phi \hat{I}_1^{-\Phi} \rangle = \frac{e^4}{\pi^3 h^2} \frac{C_0}{\gamma_0} \frac{\omega}{\gamma_0} \langle \delta G_1^\Phi \delta G_1^{-\Phi} \rangle.
\]

We also notice that in the limit $N_{ch} \gg 1$ the correlation function of the photovoltaic current $I_1^\Phi$ and the rectification current $I_1^\Phi$ vanishes [7].

First we use Eqs. [7] and [8] to analyze the magnetic field symmetry of the rectification current $I_1^\Phi$. Although in the adiabatic limit $\omega \ll \gamma_0 / h$ the rectification current $I_1^\Phi$ is symmetric with respect to magnetic field inversion ($\Phi \rightarrow -\Phi$), at higher frequencies $\omega \gg \gamma_0 / h$ the symmetry of $I_1^\Phi$ is suppressed. Indeed, the magnetic field symmetry is related to the time inversion symmetry. For a harmonic field at frequency $\omega$, the time-inversion symmetry holds only on time scales much smaller than $1 / \omega$. Transport through the system is determined by times of the order of $h / \gamma_0$ and consequently the magnetic field symmetry of the rectification current breaks if $\hbar \omega \gg \gamma_0$. We plot the ratio $S_1 = \langle \delta G_1^\Phi \delta G_1^{-\Phi} \rangle / \langle \delta G_1^\Phi \rangle^2$ as a function of...
\( \hbar \omega / \gamma_e \) in Fig. 4. \( S_1 = 1 \) represents the symmetric current \( I_1^s \) with respect to magnetic field inversion. In the adiabatic regime this symmetry originates from the Onsager symmetry of the dc conductivity, see Eq. (15) and Refs. 2, 3. As the frequency increases, \( S_1 \) vanishes, signalling the suppression of the magnetic field symmetry. Therefore, the absence of magnetic field symmetry no longer serves as a distinct feature of the photovoltaic current \( I_{ph}^p \), which allows one to distinguish \( I_{ph}^p \) and the rectification current \( I_1^r \).

We notice that the magnetic field symmetry of the dc conductance \( \delta G_0^\parallel \) is more sturdy than the symmetry of the rectification current, see Fig. 4. Particularly, at temperatures \( k_B T \gtrsim \gamma_e \), dc conductance \( \delta G_0^\parallel \) is nearly symmetric at frequencies \( \hbar \omega \lesssim k_B T \), since the dc correlation function is determined by processes on a time scale \( \hbar/k_B T \). The symmetry is not fully suppressed even at \( \omega \gg k_B T/\hbar \); the suppression depends on \( C_0/\gamma_e \).

We apply Eqs. (16) - (18) to the analysis of the experiment 3. The quantum dot used in the experiment has an area \( A \approx 0.7 \, \mu m^2 \). Relevant energy scales are the Thouless energy \( E_T \approx 160 \, \mu eV \) and the mean level spacing \( \delta_1 = \pi \gamma_e \approx 10 \, \mu eV \). The measurements were performed at the base electron temperature \( T \approx 200 \, mK \) \( (k_B T = 5.4 \gamma_e) \). From the size of conductance fluctuations without ac fluctuation on the gate, we estimate the dephasing rate \( \gamma_\phi \approx 0.2 \gamma_e \) (see 17) and thus disregard it in our quantitative analysis.

In the upper panel of Fig. 2 we show the variance of the conductance as a function of the incident power \( P = P_0 C_0/\gamma_e \) at \( \omega/2\pi = 5.56 \, GHz \) \( (\hbar \omega/\gamma_e \approx 7.2) \). We also plot the variance of the conductance calculated from Eq. (11) \( (k = 0) \) at temperature \( T = 5.4 \gamma_e/k_B \). Assuming that the ratio \( C_0/\gamma_e \) is proportional to the power \( P \) of the ac excitation applied to the gate, i.e. \( C_0/\gamma_e = P/\gamma_0 \), we rescale \( P \) to obtain the best fit of the experimental points by the curve of Eq. (11). We find \( P_0 = 9 \times 10^{-8} \, W \).

In the lower panel of Fig. 2 we show the rectification current \( I_{ph}^p \) as a function of power \( P = P_0 C_0/\gamma_e \) with \( P_0 = 2.5 \times 10^{-7} \, W \). Solid line shows variance of the photovoltaic current Eq. (11) at temperature \( T = 5.4 \gamma_e/k_B \) and frequency \( \omega = 3.1 \gamma_e/\hbar \). The dashed and dotted lines show the symmetric and antisymmetric correlators of the rectification current Eq. (11) with \( \alpha_\omega = 4.7 \hbar \omega/e \) and \( \varphi_1 = 0 \).

\[ \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I_{ph}^p \rangle \approx \langle I}_{
is no fitting parameters for the variance of the current (along vertical axis). At $C_0 \lesssim \gamma_e$, the variance of the measured current changes quadratically in $C_0/\gamma_e$, consistent with quadratic dependence on $C_0$ of the theoretical curves for $\langle \hat{I}_{ph}^2 \rangle$ and therefore our assumption that $C_0$ is proportional to the power of the ac excitation is justified. At $C_0 \gtrsim \gamma_e$ the variance of the measured current starts saturating. This saturation is expected for large power asymptote of the photovoltaic current due to spreading of the distribution function of electrons in the dot. Some deviation between the experimental points and the theoretical curve is expected due to the approximation $N_{ch} \gg 1$ used for derivation of Eq. (6) ($N_{ch} = 2$ in the experiment).

For illustration, we also plot the correlation functions of the rectification current, using Eq. (5) for $\varphi_1 = 0$ and $\alpha_\omega = 0.45h\omega/e$. For the rectification current, the saturation at large power is not expected: according to Fig. 2, $\alpha_\omega = 0$ and $\gamma_e = 83$. For comparison we plot $I_{ph} = 3.1\gamma_e$. We observe that the fluctuations of the measured current significantly exceed (by a factor $\sim 100$) the expected magnitude for the photovoltaic current, and therefore are likely due to the rectification of the bias across the dot. The low power data can be fitted by Eq. (5) with $\alpha_\omega = 4.7h\omega/e$.

The above choice for $\alpha_\omega \gtrsim (h\omega, k_B T)/e$ limits the applicability of the linear expansion Eq. (4) to small powers of the ac excitation $C_0$, such that $C_0/\gamma_e \lesssim (h\omega/e\alpha_\omega)^2$. The higher order corrections in the bias $V_\omega$ do not restore magnetic field symmetry, which is in apparent contradiction to the observed symmetry of the measured current at larger powers (at $C_0/\gamma_e \gtrsim 1$ in Fig. 3). We attribute the restoration of magnetic field symmetry to dephasing due to dot heating by the dissipative current. Increasing the power $P$ at fixed $\omega$ drives the system into the adiabatic regime since the heating makes the ratio $h\omega/(\gamma_e + \gamma_\phi)$ decrease. As shown already in Fig. 1, the rectification current is symmetric in the adiabatic regime. The assumption that $\gamma_\phi$ increases as power $P$ increases is consistent with the observed change of the correlation field for the current fluctuations, see Fig. 4 in Ref. 3.

In summary, we studied ensemble fluctuations of dc current through an open quantum dot subject to oscillating perturbation. We showed that as frequency of the perturbation increases, magnetic field symmetry of the current disappears, regardless of the mechanism of the current generation. We demonstrated that the power behavior of the current fluctuations is an important tool to distinguish effects of an ac excitation on dc current.

We thank I. Aleiner, P. Brouwer, and V. Falko for useful discussions, and M. Hanson and A. C. Gossard at UC Santa Barbara for high-quality heterostructure materials used in the experiments. The work was supported by NSF grants DMR 02-13282 and DMR 0072777, and by AFOSR grant F49620-01-1-0475.