Fractional Coulomb blockade in a coupling controlled metallic quantum dot.

O. Bitton,1, 2 A. Frydman,2 R. Berkovits,2 and D.B. Gutman2

1 Chemical Research Support department, Weizmann Institute of Science, Rehovot, Israel
2 The institute of nanotechnology and advanced materials, The Department of Physics, Bar Ilan University, Ramat Gan 52900, Israel

(Dated: June 17, 2015)

We use a novel technique to experimentally explore transport properties through a single metallic nanoparticle with variable coupling to electric leads. For strong dot-lead coupling the conductance is an oscillatory function of the gate voltage with periodicity determined by the charging energy, as expected. For weaker coupling we observe the appearance of additional multi-periodic oscillations of the conductance with the gate voltage. These harmonics correspond to a change of the charge on the dot by a fraction of an electron. This notion is supported by theoretical calculations based on dissipative action theory. Within this framework the multiple periodicity of the conductance oscillations arises due to non-pertubative instanton solutions.

PACS numbers: 72.80.Ng; 73.61.Jc; 73.40.Rw; 72.20.Ee

Transport through a quantum dot (QD) has been extensively studied in the context of two dimensional electron gas in semiconducting systems (for reviews see [1–4]). With nanotechnology advances and growing interest in electronic transport through nano particles and molecular devices it becomes important to understand the fundamental properties of metallic based QDs. A distinct property of QDs is the presence of electric conductance oscillations as a function of gate voltage, \( V_g \), that originate from the Coulomb blockade (CB) effect. Due to the strong sensitivity to the applied voltage these oscillations can be potentially utilized as building blocks of a single electron transistor.

By controlling the coupling between the dot and the leads one can drive the system from the "closed" to the "open" dot regime. The charge confined in a closed dot is quantized and one observes a well pronounced set of conductance peaks, separated by the charging energy \( E_c \). Each peak is associated with the change of total charge on the dot by one electron. On the other hand, the charge in an open dot is not quantized; Coulomb effects are suppressed and one observes a weak modulation of the conductance through the dot, with the period \( E_c \). [4, 5].

In the current work we explore the transition between these two regimes. For this we use a unique experimental method that enables us to tune the coupling between a metallic nanoparticle and the leads while controlling \( V_g \). In the intermediate regime the conductance is characterized by a multiple periodicity as a function of the gate voltage. The additional harmonics are associated with a change of the charge of the dot by a fraction of an electron.

In QDs based on low-carrier-density 2D electron gases, dot-lead coupling can be controlled by applying back gate voltages. A similar technique for controlling the coupling between leads and a high-density metallic QD doesn’t exist and modifying the coupling presents a major challenge. Several methods have been used for studying QDs based on metallic nanoparticles or other nano-objects. These include discontinuous films [6], electromigration [7], electrostatic trapping [8, 9], break-junctions [10] and angle evaporation [11]. None of these techniques provide a way to control the coupling. As a result one usually ends up with a weakly coupled dot.

We have developed a novel technique for fabricating QDs based on metallic nanoparticles, while controllably varying the coupling to leads [12, 13]. The quantum dots are chemically formed gold colloids, 30nm in diameter. Coupling to leads is achieved as follows: on a Si-SiO_2 substrate we fabricate two gold electrodes (source and drain) separated by a gap of 10 – 30nm and a perpendicular side gate electrode at a distance of 150nm. We then electrostatically connect gold colloids to the surface and use Atomic Force Microscope (AFM) nanomanipulation to "push" a desired particle to the right position between the source and drain electrodes. At this stage the dot is usually very weakly connected to the leads.
We vary the dot-lead coupling using an electrochemical method by which we deposit gold atoms on the gold electrodes to decrease the dot-lead distance [14]. During the deposition process we measure the conductance between the source and the drain and stop the process at any desired coupling. We then cool the system to T=4.2K and measure conductance as a function of gate voltage and source drain voltage, \( V_{SD} \). Further technical details can be found elsewhere [12]. An image of a dot-lead system is presented in Fig.1.

We have measured 17 samples, out of which seven showed several harmonics of \( E_c \). It turns out that probability to observe multiple harmonics depends on the dot-lead coupling, characterized by the dimensional conductivity \( g_D = (\hbar/e^2)(R_S^{-1} + R_D^{-1}) \), where \( R_S/D \) is the resistance between the dot and source/drain. Unfortunately, it is not possible to measure \( g_D \) directly from the conductivity because the dot is usually highly asymmetrically coupled. While the measured conductivity is governed by the weakly connected lead, \( g_D \) is determined by the the well connected lead. Nevertheless it is possible to extract \( g_D \) from I-V curves, such as that shown in the inset of Fig.2 [13], by fitting them to the results of Ref. [15]. Doing so we find that the coupling of all our samples is in the range \( 1 < g_D < 10 \). Fig.2 shows the number of conductance oscillation periods observed in the system as a function of coupling strength \( g_D \). It is seen that for \( g_D > 6 \) the conductance shows only a single period. Additional harmonics appear for \( g_D < 6 \) and proliferate as the system is pushed towards lower values of \( g_D \).

Conductance versus gate voltage curves \( \sigma(V_g) \) for two QDs are shown in Fig.2 panels a and b. In Fig.2a the dot is strongly coupled and the conductance oscillates with a single period corresponding to \( E_c \sim 0.8V \). In Fig.2b, on the other hand, the dot is weaker coupled and the conductance exhibits a much richer structure. The Fourier transform depicted in Fig. 2c reveals that the conductance curve is composed of 7 well defined periodicities which are identified as harmonics of the basic CB oscillation which in this case is \( \sim 1.8V \). It should be noted that the relative amplitude of the different oscillations is not monotonic with the harmonic order. In this case the second harmonic has a larger amplitude than the first and the fifth harmonic is the most prominent.

We also note that high and low harmonics are differently affected by external bias. As the bias voltage is increased, the additional harmonics are suppressed. This is demonstrated in Fig.4 which shows \( \sigma(V_g) \) for two different dots at several values of bias voltage. While for small bias voltage an extra harmonic is clearly observed in the conductance curves, at higher \( V_{SD} \) this harmonic is unmeasurable.
To get a better understanding of the behaviour of our system we use the following theoretical model. We assume that the dynamics inside the dot is chaotic [10]. The coupling of the dot to the leads is achieved by a large number of closed channels, so that the total coupling strength $g_D$ is large ($g_D \gg 1$). Because the charge inside the dot strongly fluctuates, it is convenient to use a canonically conjugated variable - phase $\phi$ [17]. The QD is thus described by a "dissipative action" [15, 17–20]. At equilibrium, at temperature $T$, it is governed by imaginary time action

$$
S = \frac{gT^2}{4} \int_0^{1/T} d\tau_1 d\tau_2 \frac{e^{i\phi(\tau_1) - i\phi(\tau_2)}}{\sinh^2 \pi T (\tau_1 - \tau_2)} + \frac{1}{4E_c} \int_0^{1/T} d\tau \dot{\phi}^2 - i q \int_0^{1/T} d\tau \dot{\phi} \dot{\phi}.
$$

(1)

Here $C$ is the capacitance of the dot that is determined by capacitive coupling to the source, drain, and the gate, $C = C_S + C_D + C_g$. The average number of electrons on the dot is

$$
q = \frac{C_S R_S - C_D R_D}{e(R_S + R_D)} V_{SD} + \frac{C_G}{e} V_g.
$$

(2)

The action (1) has non-trivial minima

$$
e^{i\phi(\tau)} = \prod_{n=1}^{|W|} \left( \frac{e^{2\pi i \tau} - z_n}{1 - z_n e^{2\pi i \tau}} \right)^{\text{sgn} W},
$$

(3)

known as Korshunov instantons [21]; $z_n$ and $\bar{z}_n$ are global variables that determine the position and the size of the instanton; $W$ is a winding number that counts the number of times the phase $\phi$ circles around the origin. Above a certain temperature (of the order $T_\ast \approx E_c \exp[-g_D]$) the instantons are rare, and they independently contribute to the conductance [20].

$$
G(T) \simeq \frac{R_D R_S}{(R_S + R_D)^2} \left[ \frac{e^2}{\hbar} \sum_{W=1}^{\infty} a_W e^{-F_W(T)} \cos(2\pi qW) \right].
$$

(4)

The first term corresponds to the topologically trivial contribution with the winding number $W = 0$. It accounts for renormalization of the Drude conductance due to the zero bias anomaly around trivial minima ($\phi$ being a constant). It results in replacement of the coupling strength by the renormalized value $g_D \to \tilde{g}_D = g_D + \ln(1 + \omega^2 t^2_c)$, controlled by the infrared energy scale $\omega = \max(T, eV)$ and $RC$ time $t_c = \frac{R_D R_S}{R_S + R_D} C$. The detailed study of zero bias anomaly in this geometry was performed in [13]. The topologically non-trivial solutions give rise to various harmonics in oscillations of conductance with the gate voltage. The instantons with the winding number $W$ correspond to a harmonic $W$ in the conductance oscillation. In the open dot limit, only the first harmonic survives, leading to weak single period oscillation [22]. As the coupling of the dot decreases, a finite number of harmonics is observed. For $g$ of the order unity an infinite number of harmonics appear with a parametrically equal magnitude, and the instanton expansion breaks down. This result merges with the one known for the strong Coulomb blockade regime [3], i.e. for $g \ll 1$. In this case the conductance is

$$
G \simeq (R_S + R_D)^{-1} \frac{\Delta E / 2T}{\sinh \Delta E / 2T}
$$

(5)

where $\Delta E = E_c (q - |q|)$ is the deviation from the degeneracy point. At $T \ll E_c$ it corresponds to the sequence of well resolved CB peaks, that in a Fourier space gives rise to a large number of harmonics with approximately same magnitude.

For the open dot limit the amplitude of harmonic number $W$ is controlled by

$$
F_W(T) \simeq \frac{\tilde{g}_W}{2} + \frac{\pi^2 T}{E_c} W^2
$$

(6)

$\tilde{g}_W \simeq g^{W+1} \psi(m)/(2\pi)$, where $\psi(m)$ is the $m$th order derivative of the digamma function. The calculation performed above is valid at equilibrium. Though Korshunov instantons can be computed within the real time Keldysh formalism [29], the non-equilibrium generalization remains to be done. On the phenomenological level, the thermal smearing out of equilibrium is accounted by [15] $F(T) \to F(T) + (2\pi/\epsilon)^2 W^2 \sum_{r=S,D} y_r(x)/R_r$, where $x_r = R_S R_D eV C/(R_S + R_D)^2$, and $y(x) = x \arctan(x) - 1/2 \ln(1 + x^2)$.

Considered as a function of $q$, the contribution of the instanton with winding number $W = 1$ is periodic with the period unity, that corresponds to the charge quantization. Indeed, changing the average number of electrons...
in the dot by one, a single periodicity in the thermodynamic potentials and transport coefficients is expected. Instantons with higher winding number give rise to fractional periodicity that corresponds to an average charge change of $1/W$ of the electron charge. In an open dot any fraction of the electron can be distributed between inside and outside of the dot, thus periodic dependences with respect to a fraction charge change are possible. We believe this picture, i.e. the occurrence of additional harmonics in the conductance is a universal property of open dots, however the weights of the harmonics are model specific. For a not-fully-chaotic dot the statistics of wave function is not universal, and there is a finite probability to find a strongly coupled states with a fraction of an electron inside the dot. Because such states give rise to the pronounced oscillation with a corresponding harmonic, one expects large sample to sample fluctuations of the harmonic strength. This may be the reason why for part of our samples the strengths of certain high harmonics is higher than of the lower ones (see Fig. 5). This view is consistent with the observed magnetic field dependence. Application of a magnetic field of the order of a flux quantum through the dot changes the single particle wave functions, thus affecting the relative magnitudes of different harmonics. Indeed, the Fourier transform shown in Fig. 5 for one of our dots reveals that the relative strengths of various harmonics oscillate with the magnetic field. One notes that the dominant harmonic changes from the first (for $B = -2T$) to the second (at $B = 0T$) and back to the first at (at $B = 3T$). The periodicity range of $5T$ corresponds to a single flux quantum penetrating the dot.

Within the dissipative action theory the influence of the temperature/bias suppresses the higher harmonics stronger than the lower ones. It results from the two effects acting together: (a) the thermal broadening of the instanton contribution, Eq. (6), is multiplied by the winding number square ($W^2 T/E_c$); (b) the zero bias anomaly leading to the logarithmic renormalization of $g_D$ is weakened at high temperatures (compared with the scale $t_e$), and the terms $g_D W$ increases with temperature. Therefore increasing temperature (or voltage) suppresses the harmonics with higher winding numbers stronger than those with the lower ones. This behavior agrees with our measurements.

To conclude, we have studied the transport through metallic quantum dots while controllably varying its coupling to the leads. We find that on the route towards fully opening, the system passes through intermediate coupling regime, in which the conductance shows multi periodic oscillations with the gate voltage. We attribute these oscillations to the charging of the dot by a fraction of an electron. This notion is supported by calculations based on the dissipative action theory.

We are grateful for useful discussions with I.S. Burmistrov, Y. Gefen and M. Titov. This research was supported by the Israeli Science Foundation (grant number 699/13 and 584/14), and German-Israeli Foundation (project 1167-165.14/2011).

[1] M.A. Kastner, Rev. Mod. Phys. 64, 849 (1992).
[2] U. Meirav and E.B. Foxman, Semicond. Sci. Technol. 11, 255 (1996).
[3] L.L. Aleiner, P.W. Brouwer, L.I. Glazman, 358, 309 (2002).
[4] Yu. Nazarov and Y. Blanter, "Quantum Transport: Introduction to Nanoscience" (University Press, Cambridge, 2009).
[5] A. Furusaki and K.A. Matveev, Phys. Rev. Lett. 75, 709 (1995).
[6] D.C. Ralph, C.T. Black, and M. Tinkham, Phys. Rev. Lett. 74, 3241 (1995).
[7] H. Park, A.K.L. Lim, A.P. Alivisatos, J. Park, and P.L. McEuen, Appl. Phys. Lett. 75, 301 (1999).
[8] A. Bezryadin, C. Dekker, G. Schmid, Appl. Phys. Lett 71, 1273 (1997).
[9] F. Kuemmeth, K.I. Bolotin, S.-F. Shi, D.C. Ralph, Nano Letters 8, No.12, 4506 (2008).
[10] M.A. Reed, C. Zhou, C.J. Muller, T.P. Burgin, and J.M. Tour, Science 278, 252 (1997).
[11] D.L. Klein, P.L. McEuen, J.E. Bowen Katari, R. Roth, and A.P. Alivisatos, Appl. Phys. Lett 68, 2574 (1996); D.L. Klein, R. Roth, A.K.L. Lim, A.P. Alivisatos, and P.L. McEuen, Nature (London) 389, 699 (1997).
[12] L. Bitton and A. Frydman, Appl. Phys. Lett 88, 113113 (2006).
[13] L. Bitton, D. B. Gutman, R. Berkovits and A. Frydman, Phys. Rev. Lett. 106, 016803 (2011).
[14] A.F. Morpurgo, C.M. Marcus, and D.B. Robinson, Appl. Phys. Lett. 74, 2084 (1999).
[15] D.S. Golubev, J. König, H. Schoeller, G. Schön, A.D. Zaikin, Phys. Rev. B 56, 15782 (1997).
[16] Y. Alhassid, Rev. Mod. Phys. 72, 895 (2000).
[17] G. Schön and A. D. Zaikin, Phys. Rep. **198**, 237 (1990).
[18] I.S. Burmistrov, A.M.M. Pruisken, Phys Rev B **81**, 085428 (2010).
[19] Yu.V. Nazarov, Phys. Rev. Lett. **82**, 1245 (1999).
[20] A. Altland, L.I. Glazman, A. Kamenev, and J.S. Meyer, Ann. Phys. (N.Y.) **321**, 2566 (2006) and references therein.
[21] S.E. Korshunov, JETP Lett. **45**, 434 (1987).
[22] G. Goeppert and H. Grabert Eur. Phys. J. B **16**, 687 (2000).
[23] M. Titov and D.B. Gutman, "Korshunov instantons in real time", to be published.
[24] X. Wang and H. Grabert, Phys. Rev. B **53**, 12621 (1996).
[25] S.V. Panyukov and A.D. Zaikin, Phys. Rev. Lett. **67**, 3168 (1991).