Long Storage Times and Control of Electrons in a Metallic Double-Dot System

Yuval Vardi, Avraham Guttman, and Israel Bar-Joseph
Department of Condensed Matter Physics, Weizmann Institute of Science, Rehovot 76100, Israel
(Dated: January 9, 2014)

In this work we investigate the dynamics of a single electron surface trap, embedded in a self-assembly metallic double-dot system. The charging and discharging of the trap by a single electron is manifested as a random telegraph signal of the current through the double-dot device. We find that we can control the duration time that an electron resides in the trap through the current that flows in the device, between fractions of a second to more than an hour, making the system attractive for quantum information operations. We suggest that the observed switching is the electrical manifestation of the optical blinking phenomenon, commonly observed in semiconductor quantum dots.

PACS numbers: 73.63.Kv, 85.35.Be, 73.23.Hk, 85.35.-p

Double quantum dot systems offer a unique opportunity for studying the rich world of quantum transport [1], and are considered promising candidates for quantum information operations [2]. Current fabrication techniques allow preparation of double-dot structures from a variety of material systems with controlled size and shape, down to a few nanometers scale [3-7]. However, the strong interaction with the underlying lattice and with the environment sets a limit on the coherence time that can be obtained in these structures [8-10]. An alternative system, in which the electrons are better isolated from the surrounding world, is an electronic trap, such as nitrogen-vacancy defect center in diamonds [11] and in silicon [12, 13]. Indeed, long coherence times are reported in such traps; however, their isolation makes the measurement and control more difficult.

Here we demonstrate how these two structures, double-dots and electronic traps, are combined to yield a hybrid system in which an electron can be stored for long durations and can be easily detected and measured. The charging and discharging of the trap by a single electron is manifested as a Random Telegraph Signal (RTS) [14, 15] of the current through the double-dot device. We find that we can control the duration that an electron resides in the trap through this current, varying it between fractions of a second to minutes. Furthermore, we show that at the Coulomb blockade region, when no current is flowing, the electron can be kept in the trap for extremely long times (> hour).

The system consists of two metallic nanoparticles (NPs) on top of a Si/SiO₂ substrate [Fig. 1(a)-(b)]. The NPs are Au 34 nm spheres, covered by a dense layer of mercaptosuccinic acid that acts as a tunnel barrier for electrical current into and from the NP. The dimer is formed by covalently linking the spheres with a short organic molecule, 4,4'-biphenyldithiol, thus forming a sub-nanometer gap between them [6-16], and placed between e-beam defined electrodes by means of electrostatic trapping [17]. The Si substrate is n-doped and used as a back gate. The measurements were done in a dilution refrigerator at a temperature of 60 mK.

Typical electrical conductance properties as a function of the back gate voltage, \( V_G \), and the source-drain voltage, \( V_{SD} \), are shown in Fig. 1(c),(e). The Coulomb blockade regime, consisting of a series of low conductance regions (diamonds) near zero \( V_{SD} \) is clearly visible. The large diamonds correspond to charging an extra electron on one of the dots, and the small ones - to charging the second dot while the first dot is already charged.

A noisy behavior is readily observed throughout the \( V_G - V_{SD} \) plane of device A [Fig. 1(c)]. We find that it corresponds to a time dependent switching behavior: Figure 1(d) shows a measurement of the current \( I \) for over 140 sec. The current abruptly switches between 0.1 nA to 0.25 nA, and remains at each conductance state ("on" and "off") for time durations \( t_0 \) and \( t_1 \), which may vary in length. Such switching behavior, commonly known as random telegraph signal, is found in almost all devices we studied, each with different contrast and time durations. This "noisy" pattern is the focus of our study.

Figures 1(e)-(f) show similar measurements of another device (device B), in which the conductance contrast is smaller and the time durations are substantially shorter. The fast dynamics of this device allows us to follow and analyze its behavior over many switching events. Figure 1(g) shows a histogram of \( t_{0,1} \) that represents more than \( 10^4 \) switching events. Clear exponential distributions are seen, from which the characteristic lifetime of these states, \( \tau_{0,1} \), can be extracted. Such exponential distributions are found in all measured devices.

The significance of these well-defined lifetimes can be realized by examining the time auto-correlation function of the measured RTS,

\[
g_2(\Delta t) = \frac{\langle \tilde{I}(t) \tilde{I}(t+\Delta t) \rangle}{\langle \tilde{I}^2(t) \rangle},
\]

where \( \tilde{I}(t) = I(t) - \langle I \rangle \), and \( \langle \cdot \rangle \) represents time average. We find that this function falls exponentially at a
where the RTS signal is clearer [Fig. 2(a)], the corresponding $g_2$ was calculated [Fig. 2(b)], and $\Gamma$ was extracted [Fig. 2(c)]. We find that $\Gamma$ is slower near the Coulomb blockade region, and becomes faster away from it. This behavior of $\Gamma$ nicely mirrors the current, $I$, in the $V_G - V_{SD}$ plane [Fig. 2(d)], indicating that the charging and discharging process of the trap is governed by the attempt rate of the flowing electrons. Figure 2(e) shows $\Gamma$ as a function of $I$ for 1500 points in the $V_G - V_{SD}$ plane. Indeed, $\Gamma$ increases linearly with the current throughout, except for the Coulomb blockade region which will be discussed later.

We find a clear clustering of the measured decay rates into three groups [Fig. 2(e)]. The physical origin of this clustering can be understood by calculating the average charging state of the two dots at each $(V_G, V_{SD})$ value [Fig. 3]. We identify three main regions: the blockade [marked as I in Fig. 2(e)], in which the charging state is $(n_1 = 1, n_2 = 1)$, the upper left region (II), in which an additional state, $(2, 1)$, contributes to the conduction, and the upper right region (III), with $(1, 0)$ state. Here, $n_i$ is the number of electrons on the $i$'th dot. The clustering of $\Gamma$ follows these charging states: the two fast switching clusters (II and III) have the same charge difference, $\delta n = n_1 - n_2$, and differ in the total charge, $\Sigma n = n_1 + n_2$, while the slow region near the blockade (region I) is characterized by $\delta n = 0$. We conclude that the trap dynamics is determined by the charging state of the two dots such that the electrostatic field between the dots, $\delta n$, acts as an effective drain-source field and the electrostatic potential, $\Sigma n$, acts as an effective gate.

By examining the conductance, $G$, we prove that the trap is indeed located between the dots near their surface. Figure 3(a) shows multiple $G - V_{SD}$ scans of device A for a fixed $V_G$ value. This device exhibits a large RTS contrast [Fig. 3(d)], and, hence, it is easier to follow its conductance properties. It is seen that each conducting state ("on" or "off") is characterized by a distinct conductance spectrum, and the system jumps randomly from one to another. Hence, by measuring the conductance spectra for various $V_G$ values we can map the location of the conductance peaks of the two states in the $V_G - V_{SD}$ plane [Fig. 3(b)]. We find that device A exhibits a rather surprising behavior, where the conductance peaks are shifted vertically with respect to each other.

Clearly, a trapped electron in the vicinity of the dimer shifts the two electrochemical potential upward, and should induce a horizontal shift of the conductance spectrum in the $V_G - V_{SD}$ plane [Fig. 3(c), top panel]. A vertical shift can only be obtained if an electron moves from one of the dots into a trap that is located between the dots. This process lowers the electrochemical potential...
opportunities to control a single electron on macroscopic devices, indicating that the trap may appear in different positions between the dots, as depicted in Fig. 3(d). Indeed, we find a combination of horizontal and vertical shift in device A are, indeed, due to a trap that is located slightly away from the central region, as shown in Fig. 3(d). We find that the high contrast and high gate peak of the two states across the $V_G - V_{SD}$ plane. The contour lines shift to opposite directions along the $V_{SD}$ axis (vertically). The solid vertical black line represents the conductance peaks in the $V_G - V_{SD}$ plane. It is seen that common gating causes a horizontal shift in the conductance spectrum (top right), while differential gating causes a vertical shift of the peaks in opposite directions (bottom right). (d) The area where the trap can be located in device A (purely differential gating, green) and B (mixing of common and differential gating, brown), as calculated in the electrostatic numerical simulation.

Figure 2. (a) RTS and (b) the corresponding time auto-correlation $g_2(t)$ for $V_{SD}$ values between 16 - 19 mV. The increase of the decay rate $\Gamma$ with voltage is clearly seen. (c) The behavior of $\Gamma$ across the $V_G - V_{SD}$ plane. The blank area at the bottom is the Coulomb blockade region, where RTS cannot be detected. The markers indicate the values at which the measurements shown in a,b were conducted. (d) The behavior of the current across the $V_G - V_{SD}$ plane, demonstrating a similar dependence as $\Gamma$ in c. (e) $\Gamma$ as a function of the current, revealing the existence of 3 distinct clusters (I-blue, II-red, III-yellow). The inset shows the location of these clusters in the $V_G - V_{SD}$ plane (same colors).

tial of the first dot by $\delta \mu_1 = -(E_C - E_{CM})/2$, and raises the other by $\delta \mu_2 = -\delta \mu_1$ [Fig. 3(c), lower panel]. Here $E_C$ and $E_{CM}$ are the individual and mutual charging energies of the dots, respectively. We conduct a full numerical calculation of the induced gating of the two dots as a function of the trap location. This allows us to locate the traps in devices A and B as shown in Fig. 3(d). We find that the high contrast and vertical shift in device A are, indeed, due to a trap that is located in a rather small area, confined to the inter-dot region. The situation is somehow different for device B. Here we find a combination of horizontal and vertical shifts, with a smaller magnitude. This is consistent with a trap that is located slightly away from the central region between the dots, as depicted in Fig. 3(d). Indeed, we find a range of relative shifts in the seven measured devices, indicating that the trap may appear in different locations near the inter-dot region, restricted to a layer close to the surface of the dots [13].

This system, a dimer with a charged trap, offers unique opportunities to control a single electron on macroscopic time scales and easily measure its state and dynamics. This control is demonstrated in Fig. 3 which shows that an electron can be trapped for extremely long times in the Coulomb blockade region. To extract $\Gamma$ in this region, where the current is very small and RTS cannot be detected, we performed the following sequence: We measured the conductance at a $V_{SD}$ value, $V_M$, outside the blockade and determine whether it is in the "on" or "off" state. We then switched the voltage into a lower value in the blockade, $V_W$, and waited for a time duration $\Delta t$ at that voltage. Finally, we switched the voltage back to $V_M$ and measured again the conductance value [see inset of Fig. 3(b)]. This allowed us to determine the conditional probability $P_{11}(\Delta t)$ to find the trap in the "on" state at time $t + \Delta t$ if it was in the same state at time $t$. Figure 4(a) shows this conditional probability for various values of $V_W$. It is seen that as we lower $V_W$ and enter the blockade region the rate at which $P_{11}$ decays with $\Delta t$ becomes slower until it is nearly flat. Extracting the decay rate, $\Gamma$, from these measurements, we find that it falls fast as we enter the Coulomb blockade regime and the occupation time of the trap exceeds a minute [Fig. 4(b)].

Figure 3. (a) Multiple conductance scans of device A as a function of $V_{SD}$ at $V_G = 160$ mV. The red and blue points depict the "on" and "off" states, respectively. In each state the spectrum consists of two peaks which are shifted relative to each other in opposite directions. (b) Contour plot of the conductance peaks of the two states across the $V_G - V_{SD}$ plane. The contour lines shift to opposite directions along the $V_{SD}$ axis (vertically). The solid vertical black line represents the measurement shown in a. (c) Diagrams of the electrochemical potentials of the leads ($\mu_L, \mu_R$) and the dots ($\mu_1, \mu_2$) for the case of common gating ($\delta \mu_1 = \delta \mu_2$, top left) and differential gating ($\delta \mu_1 = -\delta \mu_2$, bottom left). The schemes at the right show the conductance peaks in the $V_G - V_{SD}$ plane. It is seen that common gating causes a horizontal shift in the conductance spectrum (top right), while differential gating causes a vertical shift of the peaks in opposite directions (bottom right). (d) The area where the trap can be located in device A (purely differential gating, green) and B (mixing of common and differential gating, brown), as calculated in the electrostatic numerical simulation.
a powerful tool to study this important effect. calibration manifestation of optical blinking, and can be used assuggest that the RTS observed in our work is the electrical
effects is due to a trapped charge at the surface [23], and
is commonly accepted that optical blinking in these sys-
ticular the abrupt switching between two distinct states,
the relatively long time scales (order of seconds), and
similarities with the optical blinking observed in semi-
date for quantum information operations.
suggesting that the system may be an interesting candi-
dation with the environment since no current is flowing,
the voltage [17]. This isolated electron has limited interac-
\[ \Delta t \]

\[ \Delta \] 

\[ \Gamma \] 

\[ I \] 

\[ \text{log-log scale} \] 

\[ \text{linear dependence} \] 

\[ \text{peak current} \] 

\[ \text{quality factor} \] 

\[ \text{inset of b} \] 

\[ \text{Coulomb blockade regime} \] 

\[ \text{approximately} \] 

\[ \text{decays linearly with current} \] 

\[ \text{dependence on current} \] 

\[ \text{over several orders of magnitude} \] 

\[ \text{limited to a narrow area in the inter-dot region} \]

We note, however, that there is a significant difference
in the temporal statistics. While in our system the dis-
tribution of the "on" and "off" durations is exponential,
the behavior in semiconductor dots is characterized by
a power-law distribution [20]. In fact, the first quantum
dot blinking model, which attributed the phenomenon to
a trapping of carrier on the surface, predicted an expone-
dential distribution of the switching times [23]. The ori-
igin of the power-law behavior remained not fully under-
stood despite intense research in the last two decades [29].
A series of modifications to the surface trap model have
been proposed, in particular - the existence of multiple
electron traps, varying with distance and/or trap depth,
near the quantum dot surface [27]. If this is indeed the
origin of the power-law behavior, one can understand why
it is not relevant in our system, where the effective traps
are limited to a narrow area in the inter-dot region. We
suggest that measurements of the electrical conductance
of semiconductor quantum dots may be instrumental in
deciphering the lingering question of optical blinking.

We would like to thank D. Mahalu and O. Raslin
for their help in the electron beam lithography, and E.
Cohen-Hoshen and J. Sperling for fruitful discussions on
fabrication techniques.

Combining the decay rates measured in the blockade
region with the previous ones, outside the blockade [pre-
sented in Fig. 2c], we find a nice matching of the two
sets of measurements [Fig. 4c]. It is seen that $\Gamma$ de-
dpends linearly on $I$ over several orders of magnitude: the
slope of $\Gamma$ versus $I$, plotted on a log-log scale, is approx-
imately 1. We repeated this measurement with $V_W = 0$
and found no decay of the probability $P_{11}(\Delta t)$ up to
$\Delta t = 10$ minutes, implying that $\Gamma$ is much longer at this
voltage [17]. This isolated electron has limited interaction
with the environment since no current is flowing, suggesting
that the system may be an interesting candidate for quantum information operations.

The characteristics of RTS in our system have many
similarities with the optical blinking observed in semi-
ductor quantum dots [19] [20] and wires [21], in parti-
cular the abrupt switching between two distinct states,
the relatively long time scales (order of seconds), and
the current (intensity) dependence [22]. Furthermore, it
is commonly accepted that optical blinking in these sys-
tems is due to a trapped charge at the surface [23], and
there is a strong experimental evidence for a gating effect
caused by this trapped charge [24] [25]. These similarities
suggest that the RTS observed in our work is the electro-
ical manifestation of optical blinking, and can be used as
a powerful tool to study this important effect.

We note, however, that there is a significant difference

\[ \text{inset of b, measuring the current at } V_{SD} = V_M, \text{ and waiting at } V_{SD} = V_W \text{ for time duration } \Delta t. \]

\[ \text{We kept } V_M = 18 \text{ mV throughout, and varied } V_W \text{ between } 12 \text{ – } 18 \text{ mV, well into the Coulomb blockade regime. (c) } \Gamma \text{ as a function of the current in a log-log scale, inside (blue dots) and outside (red circles) the Coulomb blockade region. The blue dots are the ones presented in b, while the red ones - in Fig. 2. Remarkably, the slope of the curve is approximately 1, indicating a linear dependence of } \Gamma \text{ as a function of current for over 3 orders of magnitude.} \]

Figure 4. (a) The conditional probability $P_{11}(\Delta t)$ for the
tystem to remain in the same state after wait time of $\Delta t$ in

dious $V_{SD}$ values. (b) The corresponding decay rate $\Gamma$ as
function of $V_{SD}$. These measurements were performed as
depicted at the inset of b, measuring the current at $V_{SD} = V_M$, and waiting at $V_{SD} = V_W$ for time duration $\Delta t$. We
ke$ $V_M = 18$ mV throughout, and varied $V_W$ between
12 – 18 mV, well into the Coulomb blockade regime. (c) $\Gamma$ as a function of the current in a log-log scale, inside (blue dots) and outside (red circles) the Coulomb blockade region. The blue dots are the ones presented in b, while the red ones - in Fig. 2. Remarkably, the slope of the curve is approximately 1, indicating a linear dependence of $\Gamma$ as a function of current for over 3 orders of magnitude.

[1] W. van der Wiel, S. De Franceschi, J. Elzerman, T. Fu-
jisawa, S. Tarucha, and L. P. Kouwenhoven, Reviews of
Modern Physics 75, 1 (2002)
[2] D. Loss and D. P. DiVincenzo, Physical Review A 57,
120 (1998)
[3] J. R. Petta, A. C. Johnson, J. M. Taylor, E. A. Laird,
A. Yacoby, M. D. Lukin, C. M. Marcus, M. P. Hanson,
and A. C. Gossard, Science (New York, N.Y.) 309, 2180
(2005)
[4] Y. Hu, H. O. H. Churchill, D. J. Reilly, J. Xiang, C. M.
Lieber, and C. M. Marcus, Nature nanotechnology 2,
622 (2007)
[5] K. Todd, H.-T. Chou, S. Amasha, and D. Goldhaber-
Gordon, Nano letters 9, 416 (2009)
[6] A. Guttmann, D. Mahalu, J. Sperling, E. Cohen-Hoshen,
and I. Bar-Joseph, Applied Physics Letters 99, 063113
(2011)
[7] B. M. Maune, M. G. Borselli, B. Huang, T. D.
Ladd, P. W. Deelman, K. S. Holabird, A. Kiselev, I.
Alvarado-Rodriguez, R. S. Ross, A. E. Schmitz, M.
Sokolich, C. A. Watson, M. F. Gyure, and A. T.
Hunter, Nature 481, 344 (2012)
[8] J. R. Petta, A. C. Johnson, C. M. Marcus, M. P. Hanson,
and A. C. Gossard, Physical Review Letters 93, 186802
(2004)
[9] S. Vorotxsov, E. R. Mucciono, and H. U. Baranger, Physi-
cal Review B 71, 205322 (2005)
[10] A. V. Khatskii, D. Loss, and L. Glazman, Physical
Review Letters 88, 186802 (2002)
[11] F. Jelezko, T. Gaebel, I. Popa, A. Gruber, and J.
Wrachtrup, Physical Review Letters 92, 076401
(2004)
[12] B. E. Kane, Nature 393, 133 (1998).
[13] A. M. Tyryshkin, S. A. Lyon, A. V. Astashkin, and A. M. Raitsimring, Physical Review B 68, 193207 (2003).
[14] H. Steinberg, O. Wolf, A. Faust, A. Salant, Y. Lilach, O. Millo, and U. Banin, Nano letters 10, 2416 (2010).
[15] N. Neel, J. Kroger, and R. Berndt, Nano letters 11, 3593 (2011).
[16] T. Dadosh, Y. Gordin, R. Krahne, I. Khivrich, D. Mahalu, V. Frydman, J. Sperling, A. Yacoby, and I. Bar-Joseph, Nature 436, 677 (2005).
[17] See Supplementary information.
[18] A. M. Tyryshkin, S. A. Lyon, A. V. Astashkin, and A. M. Raitsimring, Physical Review B 68, 193207 (2003).
[19] M. Nirmal, B. O. Dabbousi, M. G. Bawendi, J. J. Macklin, J. K. Trautman, T. D. Harris, and L. E. Brus, Nature 383, 802 (1996).
[20] K. T. Shimizu, R. G. Neuhauser, C. A. Leatherdale, S. A. Empedocles, W. K. Woo, and M. G. Bawendi, Physical Review B 63, 205316 (2001).
[21] J. J. Glennon, R. Tang, W. E. Buhro, and R. a. Loomis, Nano letters 7, 3290 (2007).
[22] M. E. Pistol, P. Castrillo, D. Hessman, J. A. Prieto, and L. Samuelson, Physical Review B 59, 10725 (1999).
[23] A. L. Efros and M. Rosen, Physical Review Letters 78, 1110 (1997).
[24] K. T. Shimizu, W. K. Woo, B. R. Fisher, H. J. Eisler, and M. G. Bawendi, Physical Review Letters 89, 117401 (2002).
[25] S.-J. Park, S. Link, W. L. Miller, A. Gesquiere, and P. F. Barbara, Chemical Physics 341, 169 (2007).
[26] P. Frantsuzov, M. Kuno, B. Jankó, and R. A. Marcus, Nature Physics 4, 519 (2008).
[27] R. Verberk, A. M. van Oijen, and M. Orrit, Physical Review B 66, 233202 (2002).