論文 / 著書情報
Article / Book Information

Title	Uniform charging energy of single-electron transistors by using size-controlled Au nanoparticles
Authors	Norio Okabayashi,Kosuke Maeda,Taro Muraki,Daisuke Tanaka,Masanori Sakamoto,Toshiharu Teranishi,YUTAKA MAJIMA
Citation	Appl. Phys. Lett, Vol. 100, ,
Pub. date	2012, 1
URL	http://scitation.aip.org/content/aip/journal/apl
Copyright	Copyright (c) 2012 American Institute of Physics

AIP | Applied Physics Letters

Uniform charging energy of single-electron transistors by using size-controlled Au nanoparticles

Norio Okabayashi, Kosuke Maeda, Taro Muraki, Daisuke Tanaka, Masanori Sakamoto et al.

Citation: Appl. Phys. Lett. **100**, 033101 (2012); doi: 10.1063/1.3676191
View online: http://dx.doi.org/10.1063/1.3676191
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v100/i3
Published by the American Institute of Physics.

Uniform charging energy of single-electron transistors by using size-controlled Au nanoparticles

Norio Okabayashi,[1] Kosuke Maeda,[1] Taro Muraki,[1] Daisuke Tanaka,[2] Masanori Sakamoto,[2] Toshiharu Teranishi,[3] and Yutaka Majima[4,a)]

[1]*Materials and Structures Laboratory, Tokyo Institute of Technlogy, 4259, Nagatsuta-cho, Yokohama 226-8503, Japan and CREST, Japan Science and Technology Agency, Yokohama 226-8503, Japan*
[2]*Graduate School of Pure and Applied Sciences, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8571, Japan and CREST, Japan Science and Technology Agency, Kyoto 611-0011, Japan*
[3]*Institute for Chemical Research, Kyoto University, Gokasho, Uji-shi, Kyoto 611-0011, Japan and CREST, Japan Science and Technology Agency, Kyoto 611-0011, Japan*
[4]*Materials and Structures Laboratory, Tokyo Institute of Technology, 4259, Nagatsuta-cho, Yokohama 226-8503, Japan; CREST, Japan Science and Technology Agency, Yokohama 226-8503, Japan; and Department of Printed Electronics Engineering, Sunchon National University, Korea*

(Received 18 October 2011; accepted 19 December 2011; published online 17 January 2012)

Single-electron transistors have the potential to become next-generation nanodevices and sensors owing to their small size, low power consumption, and high charge sensitivity, where the charging energy of the devices is the most important parameter determining the operational temperature. Here, we have demonstrated that the charging energy of single-electron transistors can be controlled (48 ± 4 meV) by adopting electroless gold plating to make separation-defined nanogap electrodes and employing size-controlled chemically synthesized Au nanoparticles (5.2 ± 0.5 nm) as a Coulomb island. At this charging energy, the devices can be operated up to 160 K with on/off current ratio of 60%. © *2012 American Institute of Physics*. [doi:10.1063/1.3676191]

The continuous miniaturization of silicon-based transistors promotes research on single-electron transistors (SETs) which have a conceptually different operation principle from field-effect transistors and thus have the potential to be integrated with higher density and lower power consumption.[1] A single-electron transistor consists of two electrodes (source and drain) with a nanometer-size gap separation, a Coulomb island, and a gate electrode. When the charging energy of the system by adding one electron onto the Coulomb island is considerably larger than the thermal energy, electron tunneling is suppressed by Coulomb blockade.[2] In order to operate an SET, the charging energy should be sufficiently large compared to the thermal energy, i.e., the charging energy is the most fundamental parameter that determines the operation temperature. Although numerous SET fabrication methods have been reported thus far by using silicon technology,[3,4] carbon nanotubes,[5,6] and metal Coulomb islands,[7–13] control of the charging energy remains an important subject from a viewpoint of practical application. Here, we show that our fabrication method using size controlled nanoparticle (5.2 ± 0.5 nm) and nanogap electrodes can provide the well-controlled charging energy (48 ± 4 meV).

Our SET fabrication methods are based on a combination of electron beam lithography, electroless plating,[14–16] and adsorption of synthesized Au nanoparticles (NPs),[17] which allows us to fabricate many SETs in parallel.[17] The nanogap electrode is covered by an octanethiol ($CH_3(CH_2)_7SH$) self-assembled monolayer (SAM). Decanedithiol ($HS\text{-}(CH_2)_{10}\text{-}SH$) molecules are inserted into the defect site of the SAM or by substitution with an octanethiol molecule,[18] on which decanethiol ($CH_3(CH_2)_9SH$)-protected chemically synthesized Au NPs are adsorbed. Details of SET fabrication and electron transport measurement are described in the supplementary information.[17]

A scanning electron microscope (SEM) image of a fabricated SET and its equivalent circuit are shown in Figs. 1(a) and 1(b), respectively. Numerous bright spots on the electrodes correspond to Au NPs with a diameter of 5.2 ± 0.5 nm.[17] Between the nanogap electrodes, we see a single NP marked by an arrow, which is located closer to gate1 than gate2. When the differential conductance (dI/dV) of the SET at a temperature of 9 K is plotted as functions of the drain voltage and gate voltage, we see three cycles of Coulomb diamonds (Fig. 1(c)). Note that the current modulation is observed only for gate1 and not for gate2, because the NP cannot be electrically coupled with gate2 owing to the screening by the source and drain electrodes (Fig. 1(a)). For the same sample, periodic switching of the current by the gate voltage (Coulomb oscillation) is seen clearly at a drain bias of 40 mV (Fig. 1(e)). The drain voltage-dependent tunneling current is shown for two different gate voltages, V_{on} and V_{off} (Fig. 1(f); see Fig. 1(c)). When $V_g = V_{on}$ (V_{off}), the tunneling of electrons around the Fermi level is allowed (suppressed). By comparing the experimental and theoretical Coulomb diamonds,[19] we can evaluate C_1, C_2, and C_{g1} as 0.90 aF, 0.92 aF, and 0.032 aF(=32 zF), respectively. The estimated capacitances C_1, C_2, and C_{g1} provide a charging energy, $E_c = e^2/(2C_1 + 2C_2 + 2C_{g1}) \approx e^2/(2C_1 + 2C_2)$, of 43 meV. The residual charge (Q_0), which shifts the Coulomb diamonds along the lateral axis, is estimated to be $-0.19\,e^-$. Fitting of the drain voltage-dependent currents by the orthodox theory[20] yields R_1 of 6.3 GΩ and R_2 of 1.2 GΩ. The well accordance between experiment and theory rationalizes our assumption that the resistances R_1 and R_2 are constant regardless of the drain bias, which is consistent with the electron transport measurements for alkanethiol molecules.[21,22]

a)Electronic mail: majima@msl.titech.ac.jp.

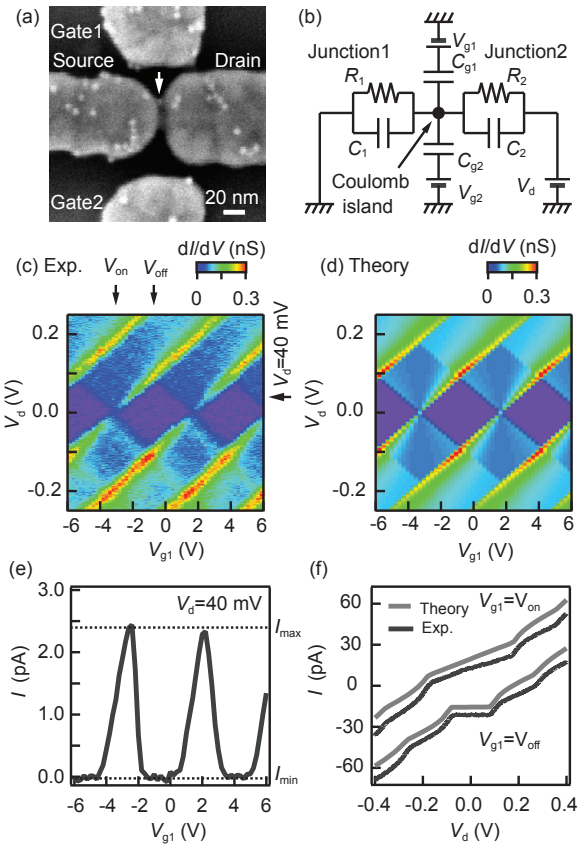

FIG. 1. (Color online) (a) SEM image of a fabricated SET, where the single NP marked by the arrow contributes to the electron transfer process. (b) Equivalent circuit of SET. (c) Experimentally observed and (d) theoretically reproduced differential conductances for the SET as functions of the drain bias and gate bias at a temperature of 9 K. (e) Coulomb oscillation at a drain bias of 40 mV. (f) Tunneling current as a function of the drain voltage (V_d) for two gate voltages. At V_{on} (V_{off}), tunneling of electrons is allowed (suppressed).

By using C_1, C_2, C_{g1}, Q_0, R_1, and R_2, we can theoretically reproduce the Coulomb diamonds (Fig. 1(d)), which agrees well with the experimental observation. In particular, the pronounced lines at the boundary of the Coulomb diamond extending from the bottom left to the top right, which originates from the large difference of resistivity between R_1 and R_2, are well reproduced. The accordance between experiment and theory clearly indicates that electron transport actually occurs via a single NP, consistent with the SEM observation.

The advantage of our fabrication method is the ability to make many SETs at once, from which we can analyze the mean value and standard deviation of SET parameters.[17] Various SETs (Figs. 2(a)–2(d)) show idealized Coulomb diamonds constituted by lines with only two slopes for each devices, i.e., electron tunneling occurs via a single NP in these devices. The ranges of the vertical axis are identical for all devices and the heights of the diamonds are almost identical. This means that the charging energies of the NPs are almost identical for all devices. Actually, the mean value of the charging energy and its standard deviation for the fabricated 9 SETs (Ref. 17) approximately originating from a single NP are 48 and 4 meV, respectively, i.e., the standard deviation is 10% of the mean value. This dispersion is more or less close to that in the diameter of the synthesized Au

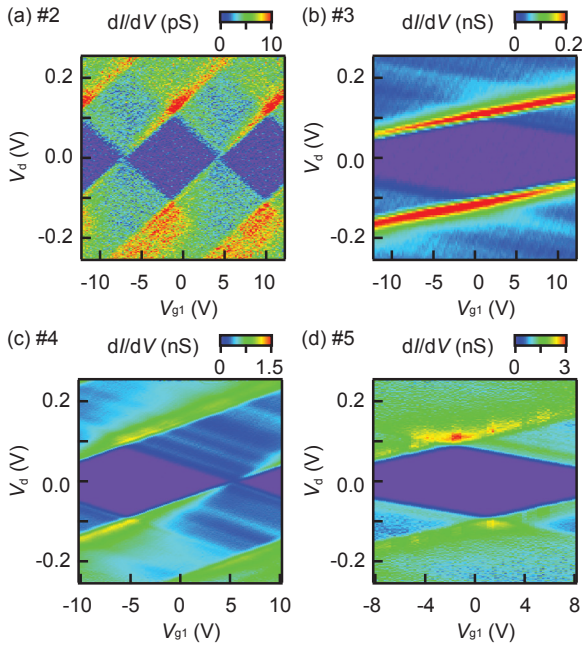

FIG. 2. (Color online) (a-d) Examples of differential conductance plots measured at 9 K that show ideal Coulomb diamonds.

NPs, i.e., the ratio of the standard deviation to the mean is 10% (0.5 nm/5.2 nm). Note that the control of the charging energy has been further confirmed by using NPs with different diameters (3.9 and 6.2 nm).

As described earlier, the charging energy of SET principally determines the operation temperature of the device. With increasing temperature, the density of states around the Fermi level becomes broad to relax the condition of Coulomb blockade (see Figs. 3(a)–3(c)). Although obscurely, the periodical Coulomb diamonds are observed up to 160 K, where the on/off ratio of the current $[(I_{max} - I_{min})/I_{max}]$ in the Coulomb

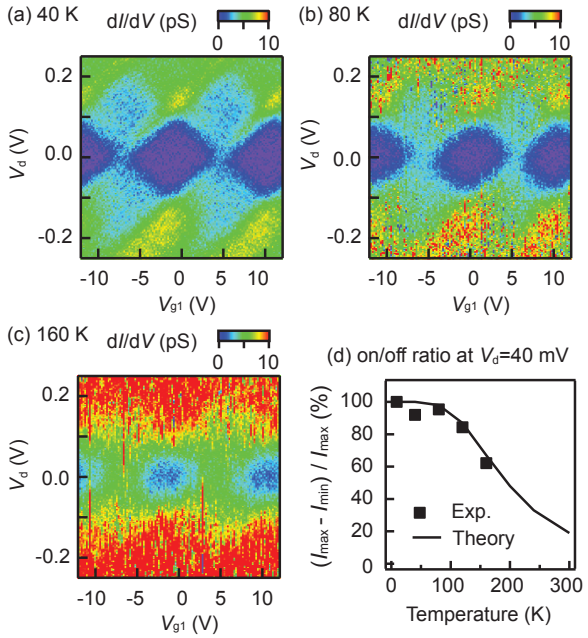

FIG. 3. (Color online) (a-c) Temperature dependent Coulomb diamonds of the device shown in Fig. 2(a). (d) Temperature dependent on/off current ratio in Coulomb oscillations.

oscillations[17] is 60%. The present finding indicates the controllability of the SET operation temperature up to 160 K by employing size-controlled NPs and nanogap electrode.

In contrast to the charging energy (or C_1, C_2), the gate capacitances (C_{g1} or C_{g2}) for the ideal 9 SETs are scattered,[17] where the mean value is 10.5 zF and the standard deviation is 8.8 zF. This result indicates that the source/drain electrodes screen the electric field between the gates and NP in various degrees depending on the position of NPs, and thus scatter the gate capacitance. The resistivities of the junctions are more diverse,[17] i.e., the geometrical averages of R_1 and R_2 are 2.9 and 1.6 GΩ, respectively and the ranges of resistivity for R_1 and R_2 derived from the geometrical standard deviation are 0.3 to 29 GΩ and 0.2 to 10 GΩ, respectively. However, given that the tunneling resistance shows exponential dependence on the tunneling distance and the decay constant of an alkanethiol molecule[23] is 1.0 $Å^{-1}$, the scattering of the resistivity is identical to only ±2.3 Å difference of the tunneling barrier.

In the present study, we have measured 236 samples of which 111 (47%) show Coulomb blockade and 125 (53%) show no tunneling current. Details of the 111 samples are as follows: 9 (3.8%) show ideal Coulomb diamonds originating from a single NP, 30 (12.7%) show multi-NP-induced Coulomb diamonds, and 72 (30.5%) show very weak modulation or very low tunneling current (less than 1 pA). Note that in the 9 approximately ideal Coulomb diamonds, satellite lines (Figs. S3(c) and S3(d)) or jagged edges of the diamonds (Figs. S3(f) and S3(g)) are observed, which is very likely caused by defects or traps near the Coulomb islands.[24,25]

In summary, we have fabricated many SETs in parallel by combining the top-down process and bottom-up process. By this advantage, we have analyzed the mean value and standard deviation of the charging energy for the fabricated 9 SETs originating from a single NP. We found that the charging energy can be well controlled, where the standard deviation is restricted as small as 10% and is as the same as that of the Au NPs size. At this charging energy, SETs can be operated up to 160 K.

We thank Ms. Michiyo Miyakawa for technical support regarding SEM. This study was partially supported by a Grant-in-Aid for Scientific Research on Innovative Areas (No. 20108011, π-Space) from the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan; by a Grant-in-Aid for Scientific Research (A) (23245028) from MEXT (T.T.); by a Grant-in-Aid for Young Scientists (B) (23710113) from MEXT (N.O.); by the Global COE Program of "Photonics Integration-Core Electronics," MEXT; by the Global COE Program of "Education and Research Center for Material Innovation," MEXT (N.O.); and by the World Class University (WCU) Program through the Ministry of Education, Science and Technology of Korea (R31-10022).

[1]K. K. Likharev, Proc. IEEE **87**, 606 (1999).
[2]S. Kano, Y. Azuma, M. Kanehara, T. Teranishi, and Y. Majima, Appl. Phys. Express **3**, 105003 (2010).
[3]L. Zhuang, L. J. Guo, and S. Y. Chou, Appl. Phys. Lett. **72**, 1205 (1998).
[4]S. J. Shin, J. J. Lee, H. J. Kang, J. B. Choi, S. R. E. Yang, Y. Takahashi, and D. G. Hasko, Nano Lett. **11**, 1591 (2011).
[5]S. J. Tans, M. H. Devoret, H. J. Dai, A. Thess, R. E. Smalley, L. J. Geerligs, and C. Dekker, Nature **386**, 474 (1997).
[6]R. Leturcq, C. Stampfer, K. Inderbitzin, L. Durrer, C. Hierold, E. Mariani, M. G. Schultz, F. von Oppen, and K. Ensslin, Nat. Phys. **5**, 327 (2009).
[7]F. Kuemmeth, K. I. Bolotin, S. F. Shi, and D. C. Ralph, Nano Lett. **8**, 4506 (2008).
[8]Y. Azuma, Y. Yasutake, K. Kono, M. Kanehara, T. Teranishi, and Y. Majima, Jpn. J. Appl. Phys. **49**, 090206 (2010).
[9]Y. Azuma, S. Suzuki, K. Maeda, N. Okabayashi, D. Tanaka, M. Sakamoto, T. Teranishi, M. R. Buitelaar, C. G. Smith, and Y. Majima, Appl. Phys. Lett. **99**, 073109 (2011).
[10]B. Gao, E. A. Osorio, K. B. Gaven, and H. S. J. van der Zant, Nanotechnology **20**, 415207 (2009).
[11]T. Nishino, R. Negishi, M. Kawao, T. Nagata, H. Ozawa, and K. Ishibashi, Nanotechnology **21**, 225301 (2010).
[12]S. I. Khondaker, K. Luo, and Z. Yao, Nanotechnology **21**, 095204 (2010).
[13]Y. Nakamura, C. D. Chen, and J. S. Tsai, Jpn. J. Appl. Phys. **35**, L1465 (1996).
[14]Y. Yasutake, K. Kono, M. Kanehara, T. Teranishi, M. R. Buitelaar, C. G. Smith, and Y. Majima, Appl. Phys. Lett. **91**, 203107 (2007).
[15]C. S. Ah, Y. J. Yun, J. S. Lee, H. J. Park, D. H. Ha, and W. S. Yun, Appl. Phys. Lett. **88**, 133116 (2006).
[16]A. Umeno and K. Hirakawa, Appl. Phys. Lett. **86**, 143103 (2005).
[17]See supplementary materials at http://dx.doi.org/10.1063/1.3676191 for the experimental details.
[18]X. H. Li, Y. Yasutake, K. Kono, M. Kanehara, T. Teranishi, and Y. Majima, Jpn. J. Appl. Phys. **48**, 04C180 (2009).
[19]Z. A. K. Durrani, *Single-Electron Devices and Circuits in Silicon* (Imperial College, London, 2010).
[20]A. E. Hanna and M. Tinkham, Phys. Rev. B **44**, 5919 (1991).
[21]N. Okabayashi, M. Paulsson, H. Ueba, Y. Konda, and T. Komeda, Phys. Rev. Lett. **104**, 077801 (2010).
[22]N. Okabayashi, M. Paulsson, H. Ueba, Y. Konda, and T. Komeda, Nano Lett. **10**, 2950 (2010).
[23]V. B. Engelkes, J. M. Beebe, and C. D. Frisbie, J. Am. Chem. Soc. **126**, 14287 (2004).
[24]H. C. George, M. Pierre, X. Jehl, A. O. Orlov, M. Sanquer, and G. L. Snider, Appl. Phys. Lett. **96**, 042114 (2010).
[25]M. Hofheinz, X. Jehl, M. Sanquer, G. Molas, M. Vinet, and S. Deleonibus, Appl. Phys. Lett. **89**, 143504 (2006).