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Nakamura Yoshiaki, Ichikawa Masakazu, Watanabe Kentaro, Hatsugai Yasuhiro

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Quantum fluctuation of tunneling current in individual Ge quantum dots induced by a single-electron transfer

Yoshiaki Nakamura\textsuperscript{a} and Masakazu Ichikawa

Quantum-Phase Electronics Center, Department of Applied Physics, Graduate School of Engineering, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan, and CREST, Japan Science and Technology Agency, Saitama 332-0012, Japan

Kentaro Watanabe and Yasuhiro Hatsugai

Department of Applied Physics, Graduate School of Engineering, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan

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A scanning tunneling microscopic study revealed quantum fluctuation of tunneling currents in individual Ge quantum dots (QDs) on SiO\textsubscript{2}/Si. This was due to the charging energy change in the QDs caused by single-electron transfer from or into the QDs. The observed electron discharging time of approximately milliseconds agreed with the propagation model of the electron wave packets from the QDs to the Si substrates by a tunneling effect rather than by passing through voids in the SiO\textsubscript{2} smaller than electron de Broglie wavelength. © 2007 American Institute of Physics.

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Self-assembled quantum dots (QDs) of group IV semiconductors have drawn much attention on account of their interesting properties such as single-electron charging effects and quantum effects.\textsuperscript{1-9} Single-electron transfer between QDs, QD-substrates, and QD-nanowires is also an interesting topic not only for its significance in scientific subjects such as quantum transport but also for its application to quantum devices because it strongly links to the origin of quantum noise,\textsuperscript{10-13} which degrades device performance. Statistical researches about the shot noise for QDs have intensively been conducted at the sample bias voltage \(V_S\) of 3–4 V and tunneling current \(I_T\) of 50–100 pA at room temperature using W tips. After STM imaging, we measured the tunneling current variation on individual QDs. We considered that the electric potential difference between QDs and substrates was negligible by considering double barriers composed of the vacuum (\(\sim\)1 nm) and the ultrathin SiO\textsubscript{2} films (\(\sim\)0.3 nm) with relative permittivity of \(\sim\)4.

Figure 1(b) shows the tunneling current variations (measured at \(V_S=+4.0\) V) on \(\sim\)2.2 nm Ge QDs indicated by the arrow in Fig. 1(a). The tunneling currents for Ge QDs were observed to fluctuate discretely when measured at relatively the Si substrates through voids formed in the SiO\textsubscript{2} films during the initial stages of Ge deposition at temperatures higher than \(\sim\)400 °C. Ge deposition at temperatures below \(\sim\)400 °C, however, formed nonepitaxial QDs that do not contact with the Si substrates because the formation of voids in ultrathin SiO\textsubscript{2} films is not sufficient at such low temperatures.\textsuperscript{1} Recent transmission electron microscopy observations confirmed that the SiO\textsubscript{2} films beneath the epitaxial Ge QDs remained, and that ultrasmall voids (\(\sim\)1 nm) existed in the SiO\textsubscript{2} films.\textsuperscript{22} STM and STS experiments were conducted at the sample bias voltage \(V_S\) of 3–4 V and tunneling current \(I_T\) of 50–100 pA at room temperature using W tips. After STM imaging, we measured the tunneling current variation on the target Ge QDs at room temperature under a weak STM z-piezo feedback loop (frequency of 0.01 Hz) whose feedback is almost negligible during the measurement period (\(\sim\)1 s) to keep a constant tip-sample distance. We performed STS experiments at the same tip-sample distance as that used during the measurement of the tunneling current variation on individual QDs. We considered that the electric potential difference between QDs and substrates was negligible by considering double barriers composed of the vacuum (\(\sim\)1 nm) and the ultrathin SiO\textsubscript{2} films (\(\sim\)0.3 nm) with relative permittivity of \(\sim\)4.

Samples cut from \(n\)-type Si (111) wafer were introduced into an ultrahigh vacuum chamber at a base pressure of \(\sim 1 \times 10^{-8}\) Pa. Si (111)-(7×7) surfaces precleaned by flashing at 1250 °C were oxidized at 600 °C for 10 min at an oxygen pressure of 2 \(\times 10^{-9}\) Pa to form ultrathin SiO\textsubscript{2} films of \(\sim 0.3\) nm in thickness.\textsuperscript{1} A Ge of 1.8 bilayers was deposited on ultrathin SiO\textsubscript{2} films at 500 °C to form epitaxial hemispherical Ge QDs (\(\sim 2\) nm) with an ultrahigh density of \(\sim 2 \times 10^{12}\) cm\textsuperscript{-2}.\textsuperscript{3,21} Epitaxial Ge QDs were contacted with

\textsuperscript{a}Electronic mail: yoshiaki@exp.t.u-tokyo.ac.jp

FIG. 1. Tunneling current variations were measured (b) on the Ge QDs indicated by arrows in STM image (a) (at \(V_S=+4\) V and \(I_T=60\) pA) taken with an almost negligible feedback loop.
become larger when QDs become smaller. A QD capacitance from an externally applied sample bias voltage is described as 

c\(\dot{C}\) = \(C_1 + C_2 = \pi d e_0 + \frac{\pi(\alpha d)^2}{4} e_0 e_{SiO_2}\),

where \(\alpha d\) is the effective size of the electric field in the SiO\(_2\) films, the SiO\(_2\) film thickness \(t_{SiO_2}\) is \(-0.3\) nm, \(e_0\) is the permittivity constant of vacuum, and \(e_{SiO_2}\) is the relative permittivity of SiO\(_2\).

FIG. 2. (a) \(V_S^e\)'s estimated from \(I_T-V_S\) curve of the 2.6 nm OD in the inset were aligned with a slope of \(-0.11\) V for the discrete tunneling current levels denoted as \(k(=1,2,\ldots)\). (b) The dot size dependence of \(E_C\) in QDs measured at \(V_S=-3\) V. The fitting curve is determined using Eq. (1) with \(a=1.3\).

high sample bias voltages (3–4 V). However, this discrete fluctuation was not observed at non-QD sites, namely, on the SiO\(_2\) films.

We considered that the discrete fluctuation of the tunneling current was caused by changes in the charging energy of QDs due to single-electron transfers from or into the QDs, where QDs were electrically charged with \(n\) electrons in the steady state under a tunneling current flow of hot electrons (\(V_S=3–4\) V) based on the theory of Averin et al.\(^5\) In this framework, the effective sample bias voltage \(V_S^e\) changes from an externally applied sample bias voltage \(V_S\) by multiples of single-electron charging energy \(E_C\). From \(I_T-V_S^e\) curves measured with STS, we estimated \(V_S^e\) values corresponding to the discrete tunneling current levels in the quantum fluctuation of the tunneling current. Figure 2(a) shows one example of \(-2.6\) nm Ge QDs. Here, the discrete levels of the tunneling current were denoted in ascending order of tunneling current as \(k(=1,2,\ldots)\). The \(V_S^e\) values estimated from the \(I_T-V_S^e\) curve shown in the inset of Fig. 2(a) were aligned for the tunneling current levels \(k\). The differences in the \(V_S^e\) values between adjacent current levels were almost constant at \(-110\) mV, as shown by the slope in Fig. 2(a). This indicates that this value corresponds to \(E_C\) in the QDs.

We investigated the dot size dependence of \(E_C\) in QDs measured from the quantum fluctuations of the tunneling current at \(V_S\) of +3.0 V, as shown in Fig. 2(b). The \(E_C\) values become larger when QDs become smaller. A QD capacitance \(C_{dot}\), having the relation of \(E_C=e/C_{dot}\) is described as the sum of the two capacitances between the tip and the dot (\(C_1\)) and between the dot and the substrate (\(C_2\)). Unlike Stranski-Krastanov islands, the present QDs were hemispherical due to the presence of the ultrathin SiO\(_2\) films.\(^1\) Thus, we applied a simple model where \(C_1\) is considered as the capacitance of a hemispherical crystal surrounded by a vacuum barrier, and \(C_2\) corresponds to a parallel-plate capacitance between the dot bases and substrates. \(C_{dot}\) of the dot capacitance with diameter \(d\) is described as

\[
C_{dot} = C_1 + C_2 = \pi d e_0 + \frac{\pi(\alpha d)^2}{4} e_0 e_{SiO_2},
\]

of \(-10\) nm, demonstrating the measurement for individual Ge QDs.

We measured the durations until a single electron gets discharged from QDs, \(\tau_d\), or charged into QDs, \(\tau_e\). For simplicity, the time taken for the transition from a lower tunneling current level \(k=1\) to a higher level \(k=2\) to occur (the time in the level \(k\) of 1) and the same in reverse (the time in the level \(k\) of 2) were defined as representative durations until a single-electron discharging \(\tau_p\) and charging, \(\tau_c\), respectively. We investigated the temperature dependence of \(\tau_p\) and \(\tau_c\) and also measured the \(\tau_d\) for nonepitaxial QDs without voids\(^1\) formed by Ge deposition at a low temperature of 350 °C [Fig. 3(a)]. The \(\tau_c\) was thought to be an apparent value due to the time resolution limit of the current amplifier (approximately sub milliseconds). On the other hand, the \(\tau_d\) was measurable and independent of the temperature. The \(\tau_d\) value for the epitaxial QDs was smaller than that for the nonepitaxial QDs.

The process of single-electron charging is potentially so fast such as the trapping of the tunneling hot electrons to the lower electronic levels in QDs, and should therefore be studied further. For single-electron discharging, the most familiar mechanism in quantum fluctuations (such as 1/\(f\) noise) is the use of thermal activation energy in the detrapping of electrons trapped at interfaces, surfaces, or defect sites.\(^10\) However, our experimental results reveal that \(\tau_p\) is independent of the temperature, therefore ruling out the electron detrapping mechanism. Also, our STS experiments revealed the lack of such defect levels near the QDs.\(^3\) We therefore considered that the discharging mechanism involves electron wave packets in QDs propagating in a nonthermal manner toward the Si substrates. The electron wave packets in epitaxial QDs are considered to pass through the voids in the SiO\(_2\) films to the underlying Si substrate on the femtosecond order, which is much faster than \(\tau_d\) (approximately milliseconds). In the present system, however, the voids in the SiO\(_2\) films between the epitaxial QDs and the Si substrate were smaller (<\(\sim\)1 nm) than the electron de Broglie wavelength (a few nm). Like optical waves, the wavelike nature of electrons means that the electron wave packets were also unable to pass through the ultrasmall voids. Electron wave packets go through the SiO\(_2\) films to the Si substrates by the tunneling effect. We simulated the time evolution of electron wave packets in the ground state of 3 nm QDs with small (0.6 nm)
and large (2.4 nm) voids in SiO$_2$ films by solving the time-dependent two-dimensional (2D) Schrödinger equation in the 2D potential $U_{\text{real}}(\mathbf{r})$ of QDs with $h$ nm voids, as shown in Fig. 3(b). The 2D potential $U_{\text{real}}(\mathbf{r})$ is composed of a 5 eV work function of Ge surrounding the 3 nm QDs and a 5 eV barrier of a 0.3 nm thick SiO$_2$ film containing $h$ nm voids. The electron ground state in QDs isolated in vacuum was used as the initial state of the electron wave packets, as shown in Fig. 3(e). Our simulation shows that electron wave packets were unable to pass through the smaller voids in SiO$_2$ films and remained trapped in QDs within several hundreds of femtoseconds [Fig. 3(e)]. This is unlike the case of larger voids, as shown by the simulation results in Fig. 3(d), where the electrons exited the QDs in 20 fs. In the electron discharging mechanism by the tunneling effect, the temperature independence of the discharging rate (1/$\tau_d$) agrees well with the present result. The difference of $\tau_d$ between epitaxial and nonepitaxial QDs can be explained by the effective tunneling barrier height difference, because the effective tunneling barrier for epitaxial QDs with subnanovoids in SiO$_2$ films is smaller than that for nonepitaxial QDs without voids.

We roughly estimated $\tau$ by the tunneling of a single electron under a one-dimensional effective potential $U_e(z_e)$ approximately described as $\int r U_{\text{real}}(\mathbf{r}) \rho(\mathbf{r}) d\mathbf{r}$, where $z_e$ is the center mass position vector, $z_e$ is the component of $\mathbf{r}_e$ in the substrates surface normal direction, and $\rho(\mathbf{r})$ is the density of the electron ground state in QDs. Using the WKB approximation, the electron transfer time $\tau$ in a semiclassical limit ($S_C \gg h$) is written as

$$\tau^{-1} = \tau_0^{-1} \exp(-S_C h), \quad (2)$$

where $S_C$ is roughly estimated as $\int dz_e 2 \sqrt{2m'} U_e(z_e)$, $\tau_0$ is the electron transfer time in the quantum limit (approximately femtoseconds), and $m'$ is the effective mass of the [111] direction (the substrate surface direction) at the conduction band minimum, $L$ point ($\sim 1.6m_0$ with electron rest mass $m_0$). We calculated $\tau$ to be $\sim 0.4$ and 60 ms for epitaxial and nonepitaxial QDs from Eq. (2), respectively. These estimates have large uncertainties of a few orders of magnitude, but they explain the correction of the electron transfer time $\tau$ by an exponential factor $e^{S_C h} \gg 1$, which is realized in the present experiment.

In conclusion, we investigated the quantum fluctuation of the tunneling current in individual Ge QDs epitaxially grown on Si (111) substrates at room temperature. The quantum fluctuation was caused by the charging energy change with the QD size dependence, resulting from single-electron transfer. The single-electron discharging was caused by the electron wave packet propagation from Ge QDs to the Si substrate by the tunneling effect with a long time constant (approximately milliseconds).

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