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

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A scanning tunneling microscopic study revealed quantum fluctuation of tunneling currents in individual Ge quantum dots (QDs) on SiO_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_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.[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,[10–13] which degrades device performance. Statistical researches about the shot noise for QDs have intensively been done.[14–20] However, the mechanism of the single-electron transfer on a scale of the electron wave packet movement has not been fully elucidated yet due to lack of microscopic studies by considering the dynamic electron wave packet movement, and not by statistical treatment in the steady state.

In the present study, we investigated the quantum fluctuation of the tunneling current in individual Ge QDs on SiO_2/Si using scanning tunneling microscopy (STM) and scanning tunneling spectroscopy (STS) at room temperature. Here, the quantum fluctuation was caused by dynamic changes in the charging energy of the Ge QDs with the QD size dependence, as a result of the single-electron transfer from or into the QDs featured by no temperature dependence. We showed the interface effect between the dot and the substrates on the single-electron transfer experimentally and theoretically by considering the electron wave packet propagation, and not by statistical treatment in the steady state.

Samples cut from n-type Si (111) wafer were introduced into an ultrahigh vacuum chamber at a base pressure of $\sim 1\times10^{-8}$ 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 2×10^{-4} Pa to form ultrathin SiO_2 films of ~0.3 nm in thickness.[1] A Ge of 1.8 bilayers was deposited on ultrathin SiO_2 films at 500 °C to form epitaxial hemispherical Ge QDs (~2 nm) with an ultrahigh density of $\sim 2\times10^{12}$ cm^{-2}.[3,21] Epitaxial Ge QDs were contacted with the Si substrates through voids formed in the SiO_2 films during the initial stages of Ge deposition at temperatures higher than ~400 °C. Ge deposition at temperatures below ~400 °C, however, formed nonepitaxial QDs that do not contact with the Si substrates because the formation of voids in ultrathin SiO_2 films is not sufficient at such low temperatures.[1] Recent transmission electron microscopy observations confirmed that the SiO_2 films beneath the epitaxial Ge QDs remained, and that ultrasmall voids (<~1 nm) existed in the SiO_2 films.[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 (<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 (~1 nm) and the ultrathin SiO_2 films (~0.3 nm) with relative permittivity of ~4.

Figure 1(b) shows the tunneling current variations (measured at V_S=+4.0 V) on ~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

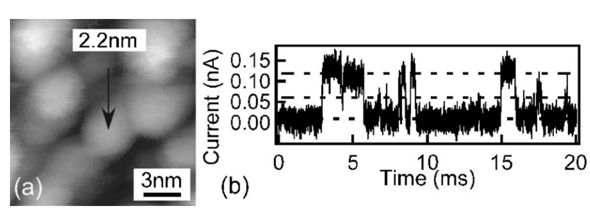

FIG. 1. Tunneling current variations were measured (b) on the Ge QDs indicated by arrows in STM image (a) (at V_S=+4 V and I_T=60 pA) taken with an almost negligible feedback loop.

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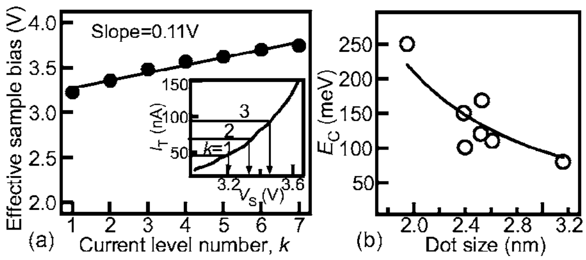

FIG. 2. (a) V_S^e's estimated from I_T-V_S curve of the 2.6 nm OD in the inset were aligned with a slope of ~0.11 V for the discrete tunneling current levels denoted as $k(=1,2,\ldots)$. (b) The dot size dependence of E_C in QDs measured at $V_S=+3$ V. The fitting curve is determined using Eq. (1) with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

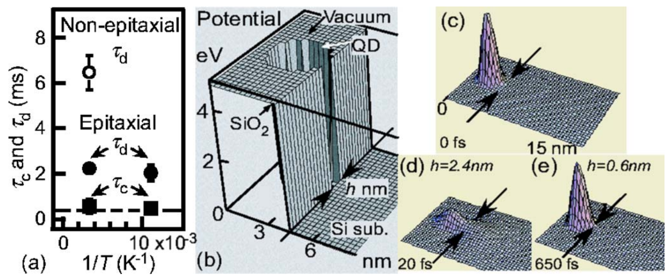

FIG. 3. (Color online) (a) Temperature dependence of the time taken until electron charging τ_c (squares) and discharging τ_d (filled circles) for epitaxial QDs. The duration τ_d for nonepitaxial QDs was plotted using open circles. The dotted line indicates the measuring limit. (b) 2D real potential of 3 nm QDs with h nm voids in a 0.3 nm thick SiO_2 film. Potential height is 5 eV. (c) The density of the electron wave packet in the ground state of 3 nm QDs isolated in vacuum. The density of electrons evolved from the initial state in (c) in (d) 20 fs in the potential with 2.4 nm voids, and in (e) 650 fs in the potential with 0.6 nm voids.

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.*[6] 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 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 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 $E_C=e/C_{\mathrm{dot}}$, 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

$$C_{\mathrm{dot}} = C_1 + C_2 = \pi d \epsilon_0 + \frac{\pi (ad)^2}{4 t_{\mathrm{SiO}_2}} \epsilon_0 \epsilon_{\mathrm{SiO}_2}, \quad (1)$$

where ad 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, ε_0 is the permittivity constant of vacuum, and $\varepsilon_{\mathrm{SiO}_2}$ is the relative permittivity of SiO_2. Figure 2(b) reveals the curve of best fit of Eq. (1) with an adjustable parameter a of 1.3. This indicated that the spatial expansion of the electric field ad ranged from 2 to 4 nm, which is smaller than the dot-dot average distance of ~10 nm, demonstrating the measurement for individual Ge QDs.

We measured the durations until a single electron gets discharged from QDs, τ_d, or charged into QDs, τ_c. 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 τ_d, and charging, τ_c, respectively. We investigated the temperature dependence of τ_d and τ_c, and also measured the τ_d for nonepitaxial QDs without voids[1] formed by Ge deposition at a low temperature of 350 °C [Fig. 3(a)]. The τ_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 τ_d was measurable and independent of the temperature. The τ_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 τ_d 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 τ_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 (<~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 $U_{\mathrm{real}}(\mathbf{r})$ of QDs with h nm voids, as shown in Fig. 3(b). The 2D potential $U_{\mathrm{real}}(\mathbf{r})$ 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(c). 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 τ_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 τ by the tunneling of a single electron under a one-dimensional effective potential $U_e(z_c)$ approximately described as $\int_{\mathbf{r}'} U_{\mathrm{real}}(\mathbf{r})\rho(\mathbf{r}-\mathbf{r}_c)d\mathbf{r}$, where $\mathbf{r}_c$ is the center mass position vector, z_c is the component of $\mathbf{r}_c$ 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 τ in a semiclassical limit $(S_C \gg \hbar)$ is written as

$$\tau^{-1} = \tau_Q^{-1} \exp(-S_C/\hbar), \tag{2}$$

where S_C is roughly estimated as $\int dz_c 2\sqrt{2m^* U_e(z_c)}$, τ_Q 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 $(\sim 1.6 m_0$ with electron rest mass $m_0)$. We calculated τ to be $\sim$0.4 and 60 ms for epitaxial and nonepitaxial QDs from Eq. (2), respectively. These estimations have large uncertainties of a few orders of magnitude, but they explain the correction of the electron transfer time τ by an exponential factor $e^{S_C/\hbar} \gg 1$, 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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