

Development of Novel Functional Si-based Devices Using Self-assembled Nanostructures for Multivalued Memory Operation, Ultimate Photo-sensing and Molecular Recognition

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1. Research Target

The serious limitations in down-sizing conventional MOS devices motivate us to develop novel functional devices operating with a few electrons and a few photons by means of an introduction of well-defined Si nanostructures into the MOS devices.

In this project, for the advanced information processing with a few electrons and photons, we focus attention on the device application of unique physical properties of Si nanocrystals associated with quantum size effects [1-2] and coulomb brocade [3] and have intensively studied MOS-FETs with quantum dots (QDs) as a floating gate [4,5] which operate even at room temperature and low voltages. Based on a deep understanding of the multivalued capability of the QDs-floating-gate, we also plan to design and fabricate optically or chemically coupling QDs-floating-gate MOSFETs. For the optically coupling devices, the gate stack structures consisting of a semitransparent metal gate, a gate dielectric with a high dielectric constant (high-k), a QDs-floating gate and the bottom tunneling oxide will be investigated to achieve operations critical to the input of a few photons. And for the chemically coupling devices, porous metal gate will be combined with a high-k dielectric as control gate insulator to selectively adsorb specific molecules on the high-k gate dielectric.

In addition, from viewpoints of the synthesis of new tailor-made electronic materials, we will extend our research work to three dimensionally stacked structures of Si-based dots with a high areal density to control unique transport properties originating from electronic coupling among neighboring dots.

Major research issues are as follows:

1. Development of process technologies to control precisely the dot size, its distribution and position and fabrication of well-ordered array and high density stacked structures of dots
2. Characterization of carrier transport through coupling dots
3. Control of energy band structures with an introduction of Ge core to Si dots
4. Valence control with impurity doping to Si-based dots
5. Fabrication and characterization of QDs-floating-gate MOSFETs for multivalued operation, high photosensitive performance and molecular recognition

2. Research Results

2.1 Self-assembling Formation of Si-QDs by Low Pressure CVD

We have demonstrated that hemispherical single-crystalline Si-QDs with a fairly uniform size distribution and a high areal density ($\sim 10^{12} \text{cm}^{-2}$) can be spontaneously formed on thermally-grown SiO_2 layers by controlling the early stages of LPCVD using a SiH_4 gas [6]. The temperature dependences of the areal dot density and the dot size in the early stages of LPCVD in the range of 560-700°C have shown that, for the dot formation on clean as-grown SiO_2 , the Si-O bond breaking plays a role in the creation of nucleation sites and the dot size is rate-limited by the SiH_4 decomposition on Si and Si cohesive action. In addition, it has been found that surface Si-OH bonds if any act as reactive sites during LPCVD to efficiently promote the dot formation. Also, the SiH_4 pressure dependence of the areal dot density at 560°C show that, in the pressure region below 0.1 Torr, a reduction in the SiH_4 pressure causes a marked decrease in the dot density, especially on as-grown SiO_2 , suggesting that the thermal decomposition of small Si clusters before reaching the critical size for stable nucleation growth becomes significant under a low SiH_4 flux. The result indicates that, to obtain high selectivity of Si dot nucleation between as-grown and OH-terminated surfaces, the suppression of the spontaneous nucleation on as-grown SiO_2 is of great importance and the control of the SiH_4 pressure during LPCVD is one of critical factors. The formation of spherical Si QDs with a Ge core is also succeeded by controlling the selective deposition conditions in LPCVD using alternately SiH_4 and GeH_4 .

2.2 Positioning of Si-QDs on SiO_2

Based on the better understanding of the Si dot formation mechanism, the spatial control of OH termination on SiO_2 leads into positioning the nucleation site. In fact, the chemical or electrochemical surface modification on a nanometer scale by using AFM/STM probe techniques enables us to make regular arrays of Si dots as shown in Fig. 1, where the as-grown SiO_2 surface just before LPCVD was exposed to the electron beam from the a Pt (20%Ir) STM tip in H_2 ambient of $1 \times 10^{-5} \text{Pa}$ [7]. Since H atoms are adsorbed on the clean Pt-tip surface through dissociative adsorption even at room temperature and emitted under a high electric field between the tip and the sample surface, reac-

tive sites such as Si-H and Si-OH bonds are formed efficiently.

2.3 Evaluation of Charged States in Si-QDs

We have demonstrated that charges retained in the single Si dot covered with ultrathin SiO₂ are directly quantified from the change in the surface potential of Si dots induced by electron injection or emission through ultrathin SiO₂ as measured with an AMF/Kelvin probe technique [8]. For the Si dots in the range of 4.3-13nm in dot height, stable retention of single electron in each dot on 4nm-thick SiO₂ was confirmed from the comparison between measured surface potential changes on the charged dots with the calculated results in the equivalent circuit mode. For Si-QDs with a Ge core, we have also found that electrons and holes are retained in the Si clad and the Ge core, respectively as predictive in the type II band discontinuity between the Si clad and the Ge core.

2.4 Characteristics of MOS Capacitors with Si-QDs as a Floating Gate

Capacitance-voltage (C-V) and current-voltage (I-V) characteristics of Si QDs floating gate MOS capacitors exhibit unique hystereses which arise from charging and discharging in the Si QDs through the bottom tunnel oxide as shown in Fig. 2. For the case on p-Si(100) with an acceptor concentration of $3 \times 10^{16} \text{cm}^{-3}$, the C-V curve measured from -3V to 3V is almost identical to the C-V curves for the case of uncharged Si-QDs floating gate. Similarly, the C-V curve measured from +3V to -3V for the case on n-Si(100) with a donor concentration of $1 \times 10^{15} \text{cm}^{-3}$, is almost identical to the C-V curve for the uncharged floating gate. In both cases, a flat-band voltage shift of $\sim 0.27\text{V}$ is observed and the capacitance peak, which is due to the emission of remaining charges to the substrate as seen in I-V characteristics, is measured around the corresponding flat-band voltage, namely the voltage separation of the peaks agrees well with the value expected from the difference in the Fermi level between p-Si(100) and n-Si(100). The results indicate that the dots act as memory nodes. In other words, we can rule out the contribution of traps with specific energy levels to the measured C-V hystereses. I-V

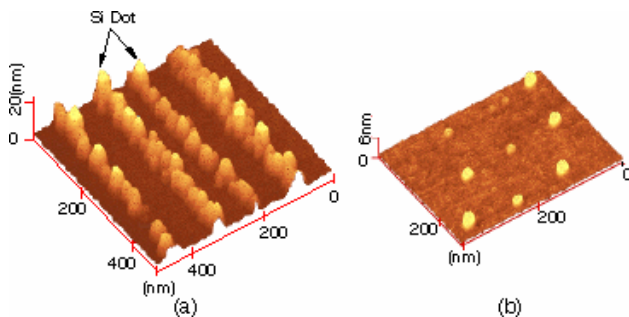


Fig. 1 AFM image obtained after Si dot formation at 560°C on the SiO₂ surface modified with spot and densely lined patterns by the STM tip. In the STM surface modification prior to LPCVD, the tip bias was applied at -10V with respect to the substrate.

characteristics shows multiple-step electron charging (or discharging) characteristics of the Si-dot floating gate are observable, suggesting that Coulombic force arising from charged dots efficiently suppresses the electron charging of neighboring neutral dots. When the floating gate consists of double-stacked dots instead of a single dot layer, the retention characteristics are improved significantly with keeping the tunneling oxide thickness constant for a high writing speed as represented in Fig. 3.

2.5 Characteristics of Si-QDs Floating Gate nMOSFETs

Based on the characteristics of MOS capacitors with the Si-QDs floating gate, n-MOSFETs with adoubly-stacked Si-QDs floating gate. The drain current-gate voltage (I_d - V_g) curves show characteristics hysteresis arising from electron charging and discharging of the Si-QDs floating gate (Fig. 4). Distinct current bumps, which were observed in ramping up the gate voltage after complete discharging at -4V, indicate the multi-step electron charging to the Si-QDs floating gate caused by the Coulomb blockade effect. Considering that, in the region of 0-0.5V, a new current component emerges with increasing sweep rate, four steps of electron injection to the Si-QDs floating gate with an areal dot density of $\sim 6 \times 10^{11} \text{cm}^{-2}$ are confirmed. In addition, the threshold voltage shift at each charging steps is not equal to that of the next charging step and slightly increase with progressive electron charging. This implies

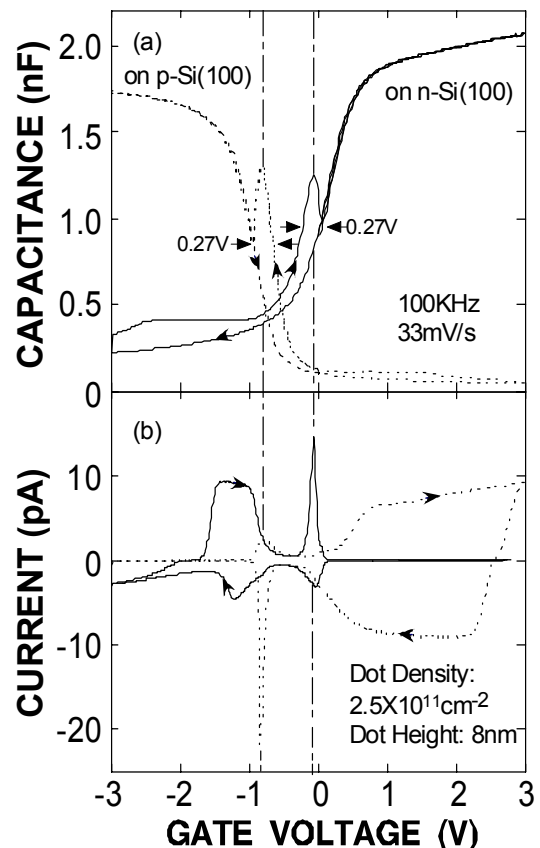


Fig. 2 Capacitance-voltage (a) and current-voltage (b) characteristics of Si-QDs floating gate MOS capacitors fabricated on p-Si(100) and n-Si(100).

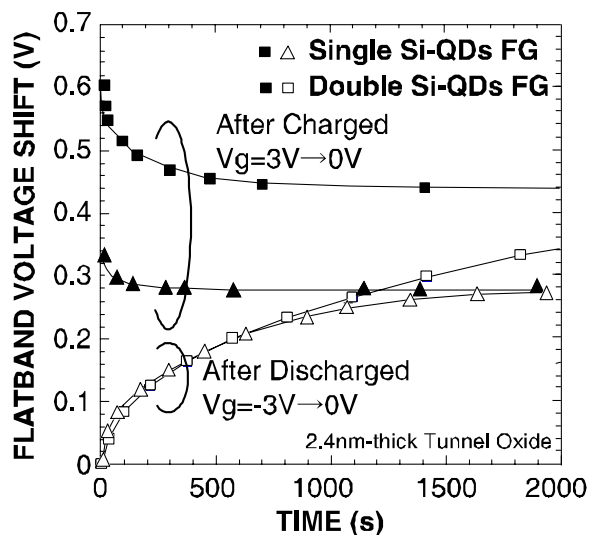


Fig. 3 Retention characteristics of MOS capacitors with the floating gate consists of double-stacked and a single-layer Si QDs.

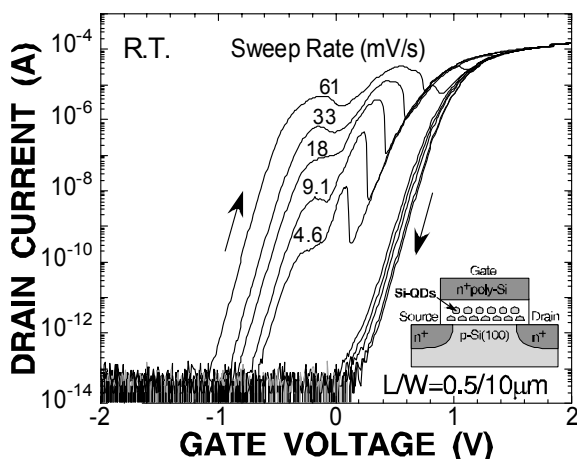


Fig. 4 Drain current vs gate voltage characteristics of a Si-QDs floating gate MOSFET, which were measured after fully discharged at a gate bias of $-4V$. The drain voltage was $50mV$. The voltage sweep rate was changed in the range from 4.6 to $61mV/s$. A cross-sectional view of a Si-QDs floating gate MOSFET is illustrated in the inset.

that the Coulomb interaction from the neighboring charged QDs play a subsidiary role on the electron tunneling from the channel to the Si-QDs. In other words, this can be interpreted in terms that the charging energy of the Si-QDs depends on not only charged states of individual Si QDs but also charged states of neighboring QDs. In general, since the charging time of the Si-QDs floating gate strongly depends on the gate voltage, faster the sweep rate is, higher the gate voltage for the charge injection becomes. In fact, with increasing sweep rate, the current bumps appear at higher gate voltages. Besides, the results of Fig. 4 implies that the charging voltage is affected by the redistribution of electrons in the Si-QDs floating gate during the gate voltage sweep as discussed later. Multi-step electron injection

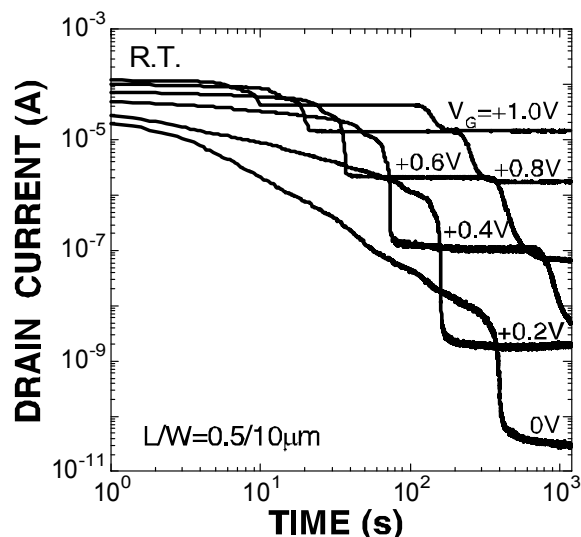


Fig. 5 Temporal change in drain current at various gate biases and a drain voltage of $50mV$ after complete discharging of a Si-QDs floating gate at a gate voltage of $-4V$.

to the Si-QDs floating gate is also clearly seen in the temporal change in the drain current (I_d-t) at constant gate biases after complete discharging of the Si-QDs floating gate (Fig. 5). The threshold voltage after each current step corresponds to that of the scanned I_d-V_g characteristics. The distinct metastable state, in which the drain current is almost constant with respect to holding time, indicate that the total amount of effective charge in the Si-QDs floating gate remains unchanged in each of the metastable states. This result suggests that electron injected in the Si-QDs floating gate redistribution during each metastable state to reduce the effect of the Coulomb interaction among the charged QDs, namely to decrease the charging energy of QDs. The temperature dependence of I_d-t characteristics measured at a fixed gate voltage show that a decrease in temperature decelerates the electron charging and prolongs the metastable states, suggesting that the tunneling probability is increased with temperature for redistribution of electrons in the Si-QDs floating gate (Fig. 6). When the charging time (Δt_1 and Δt_2) and metastable time (Δt_3) are defined with a linearly extrapolation method and their reciprocal values are summarized in Arrhenius plots, the activation energy determined from the slope of the Arrhenius plot is found to be in the range of $0.23-0.31eV$ (Fig. 7). Based on the estimation of the sum of quantized and charging energies for QDs, we found that the obtained activation energy is almost equal to the energy separation between states for tunneling in QDs. It is likely that the electron tunneling between different energy states plays an important role on the observed multiple-step charging characteristics.

The influence of light irradiation on electron injection characteristics in Si-QDs floating gate has also been studied.

The temporal changes of drain current (I_d-t) measured at constant gates in dark and under light irradiation were

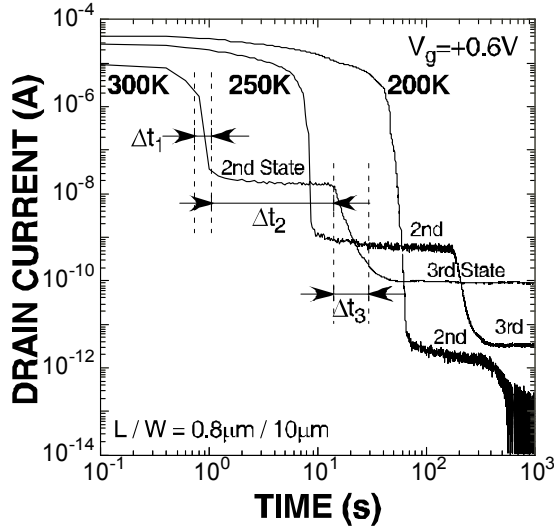


Fig. 6 Temperature dependence of I_d - t characteristics at a gate bias of 0.6V after complete discharging under the same condition of Figs 4 and 5. The drain voltage was 50mV. The definition of characteristics time (Δt_1 , Δt_2 and Δt_3) is demonstrated in the curve at 300K.

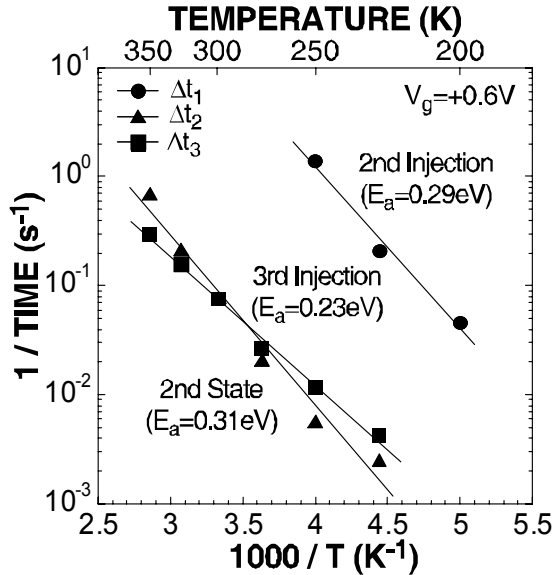


Fig. 7 Arrhenius plots of 2nd injection (Δt_1) and 2nd state (Δt_2) and 3rd injection (Δt_3) times from I_d - t characteristics.

compared as shown in Fig. 8. Although the current level is increased by 1.59eV light irradiation due to the photogenerated electron contribution, multistep electron charging of the Si-QDs floating gate and a metastable state, in which a slight decrease in the drain current is observable, are still clearly observed as in the case measured in dark. Obviously, 1.59eV light irradiation promotes electron injection to the Si-QDs and reduces the period of the metastable charged state. Notice that when the light irradiation was turned off in the stable state achieved under light irradiation, the drain current coincides with the current level of the stable state obtained in dark. This indicates that the same amount of charges were injected finally into the

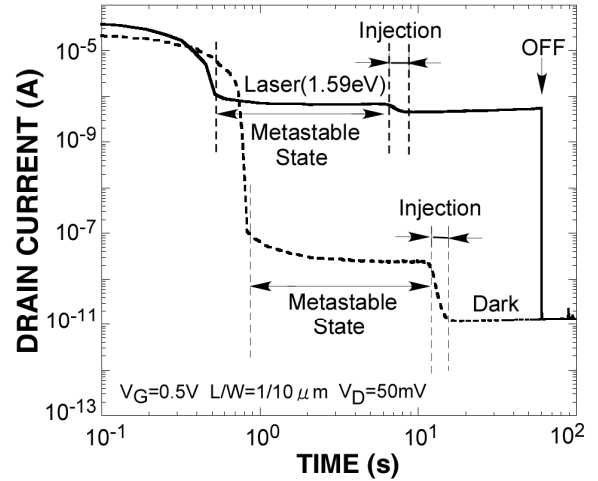


Fig. 8 Temporal changes in the drain current measured at $V_g = 0.5V$ under irradiation of 780nm (1.59eV) light and dark condition after complete discharging of the Si-QDs floating gate at $V_g = -4V$. Electron charging to the Si-QDs floating gate causes the threshold voltage shift resulting in the decrease in I_d .

Si-QDs floating gate in both cases under light irradiation and in dark condition. In other words, no excess electrons over a thermally equivalent level in dark is not injected to the dots under this light irradiation, which implies sufficient energy separation between the charged state and the next. We also found that the electron injection speed at the transition from the metastable charged state to the finally stable charged state is unlikely to be accelerated with increasing photon flux (Fig. 9) and in the photon energy (Fig. 10) although the time to the final stable state becomes shorter with higher photon flux and/or higher photon energy. The result implies that the light irradiation mainly accelerates the temporal change in the charging during the metastable charged states to trigger the transition to the final stable state. During the I_d - t measurement, when light irradiation was turned off in the early stages of metastable charged states, the drain current quickly decreases to some current level once and recovered partly due to the electron emission from the dots and then followed by the temporal change in dark (Fig. 11). Since the electron emission becomes hardly observed with the time spending in metastable states, the redistribution of electrons in the Si-QDs proceeds during the metastable charged state to further electron injection. Based on these results, we can suggest that the light irradiation plays an important role on the redistribution of electrons in the Si-QDs floating gate. Considering the fact that, by irradiation of photons with a sub-gap energy ($0.8eV$, $7 \times 10^{17} cm^{-2} s^{-1}$), no significant promotion in electron charging is observable, the contribution of the photoexcitation effect of injected electrons in the Si-QDs to the electron redistribution in the floating gate can be ruled out. The generation of hot electrons in channel with visible light irradiation is thought to be a crucial factor for the shorten metastable state, in which the electron tunneling

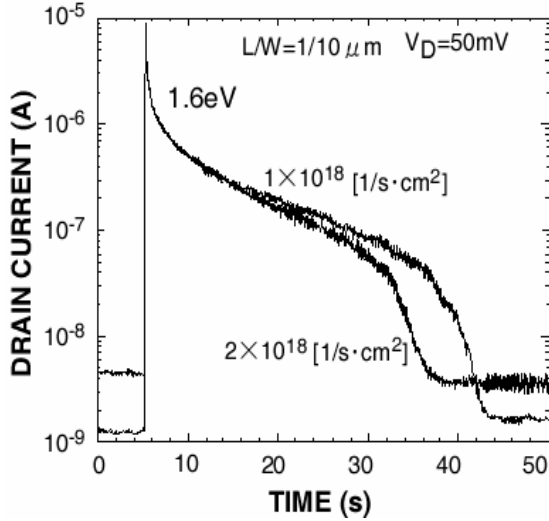


Fig. 9 The temporal changes in the drain current measured at $V_G=0V$ under 1.59eV light irradiation with photon fluxes of 1×10^{18} and 2×10^{18} cm^{-2}/s after complete discharging of the Si-QDs floating gate at $V_g=-4V$.

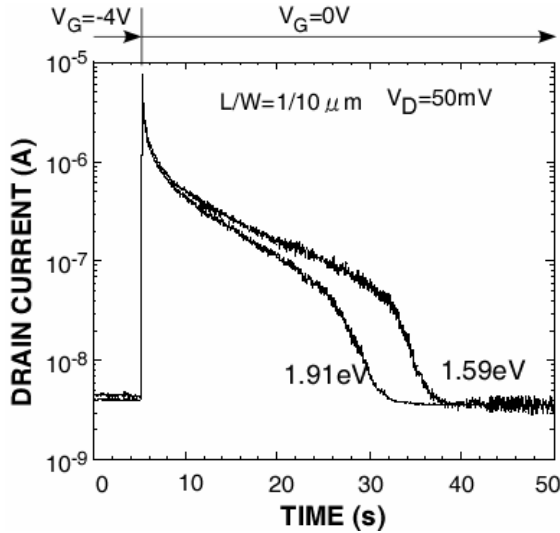


Fig. 10 The temporal changes in the drain current measured at $V_g=0V$ under light irradiation of 1.59eV and 1.91eV photons with the same flux after complete discharging of the Si-QDs floating gate at $V_g=-4V$.

seems to be controlled by the charging and quantized energy in Si-QDs.

3. Summary

We have prepared nanometer-size Si dots with and without Ge core in a self-assembling manner by controlling the early stages in low pressure chemical vapor deposition on thermally-grown SiO_2 . We have evaluated the surface potential change of each of Si dots due to charging or discharging one electron or a few by an AFM/Kelvin probe technique and demonstrated that, for dots consisting of Si clad and Ge core, electrons are stably stored in Si clad

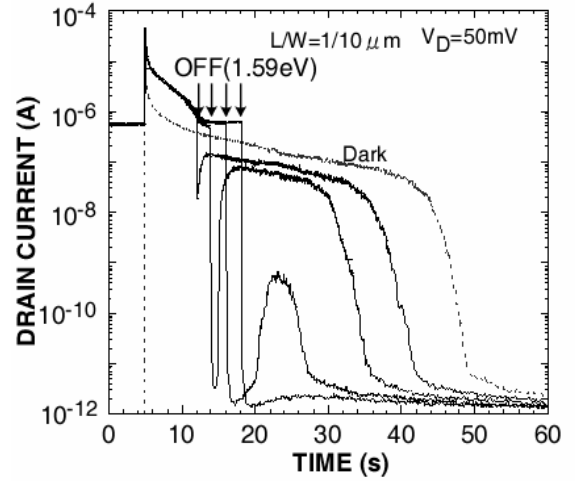


Fig. 11 Temporal change in the drain current, which was measured at $V_g=0V$ when the 1.59eV light irradiation was turned off at various time as indicated by arrows. The result obtained without light irradiation anytime is also shown as a reference.

while holes in Ge core.

Multiple-step electronic charging and discharging characteristics of Si quantum-dots (Si-QDs) floating gates in the MOS capacitors and MOSFETs have been studied in the temperature range of 200 ~ 350K. The temporal change in the drain current at a constant gate bias after complete discharging in the Si-QDs floating gate shows specific stepwise reductions accompanied with metastable states in which the drain current also remain unchanged with time until the next quick current reduction. The result indicates that, in the metastable states, injected electrons redistribute in the Si-QDs floating gate with keeping the effective total charge density in the floating gate, and suggests that the Coulomb interaction among electrons stored in neighboring dots plays an important role on the stepwise behavior in electron charging. From the slope of Arrhenius plots of the time for both each electron injection and metastable state, it is likely that a thermal activation process with an energy of 0.3eV is involved in the electron charging to the Si-QDs floating gate. The activation energy (0.3eV) suggests the electron tunneling between different energy states among neighboring Si-QDs, considering the charging energy and quantization energy in Si-QDs. Also, we have demonstrated that the electron charging of the Si-QDs floating gate is accelerated by visible light irradiation and the accelerated charging is mainly attributable to the promotion of electron redistribution in the metastable charged states.

4. Future Research Issues in the COE Program

To further improve the performance of the Si-QDs floating gate MOSFETs, we focus on the precise control of the dot size distribution and the optimization of the dot stacked structure, especially the oxide thickness between the dots. In addition, we will extend our research to

doping control of ionized impurities in Si-based dots for well-discrete charged states. Furthermore, we will fabricate photosensitive functional devices with the stacked Si QDs floating gate for optical interconnect.

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5. Published Papers and Presentations in International Conferences

[I] Published Papers on This Research

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