

4-2 Single Molecular Electronics

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In recent years, nano-technology may lead to a new way to electronics and many novel electronic devices in the field of information and communications technology. One of these nano-devices, single electron tunneling device using small particles in their systems have been successfully prepared, high-density memory devices, high-speed low power switching devices, high-sensitive electrometer devices and others will be produced in near future. However these are not sufficient. Using an organic molecule as a coulomb island electrode, it is possible to use a potential ability of the molecules. In the present paper, we fabricate the some types of organic SET device, using organic and inorganic insulator, and ultra-narrow nano-gap electrodes, and investigate the electrical properties of these single electron tunneling devices with or without light irradiation. We are making effort to adding specific optical and electronic properties coming from individual organic molecules to our single electron tunneling devices.

Keywords

Molecular electronics, Single electron tunneling device, Organic molecule, Light gate device, Nano-gap electrodes

1 Introduction

1.1 Organic Molecular Electronics

In the application of organic molecules to electronics, passive materials such as insulating materials have been the main streams in the study. These materials have been very important in the electronics but did not constitute the main stream in the characteristics of devices. Also, in the technique and the materials of the so-called “solid-state electronics”, which support current information system, organic molecules have been used in electronic parts, electronic insulating materials and coating materials as the materials of devices with special features in insulating and dielectric functions.

Under such circumstances, several decades have elapsed already since organic molecular electronics have been proposed and have become the object of the study with a basic concept to apply organic molecules in active

electric materials (e.g. rectifying device, transistor device, etc.) by using organic molecules as materials. The organic molecular electronic device as initially proposed was a device to provide electronic functions (such as rectifying function and transistor function) in a synthesized single molecule [1]. With such concept, it was tried to break through the limits of conventional Si device. At that time, it would have been impossible to actually prepare a device consisting of a single molecule and to verify its function. This was because there were technical problems in synthesizing a relatively large molecule, which has an electronic functional group in a molecule, from the viewpoint of molecular synthesis. Also, it was because there were problems of electronic access or contact to a single molecule in ultra-fine size of nano or sub-nano scale from external environment such as electronic measuring instruments and devices. For this reason, the study of organic electronics in earlier stage

has been mostly concentrated on the study based on molecular aggregate, which is relatively easy to produce. From the studies on various types of materials, devices and interface, a number of organic devices for practical use such as liquid crystal, EL device, etc. have been prepared. In recent years, a wide variety of features such as flexibility, light-weight property, economic feasibility, diversity, etc., which are characteristic of functions of organic materials, have been added to these practical devices and studies have been advancing. As a result, these features are now important factors in the manufacture of future organic devices. The studies are now flourishing, and it is attempted to produce the devices by achieving the functions of molecular crystals, molecular thin-film and macromolecular thin-film in the regions of ultra-fine size and on nano level. Emphasis is put on flexibility, e.g. in the development of organic FET, for which studies are made to achieve practical use. It is generally said that the control of physical properties on nano level and molecular level is important for the improvement of performance characteristics.

1.2 Single Molecular Electronics

The techniques to prepare organic molecular film in ultra-fine size (top-down) or to organize organic molecules (bottom-up) and the development of tools to directly evaluate and to have access to molecules such as STM, AFM, etc. has been advancing. Now, the basis to create "single molecular device" to provide active electronic function to a single molecule, as initially proposed for organic molecular electronics, has been consolidated to full extent although it is still in the stage of research. In fact, several studies [2]-[4] for the evaluation of electrical properties of single molecule have appeared. In future, it should be tried to apply the results of these studies for the research to prepare single molecular device, which represents the true aspect of the initial organic molecular electronics, without terminating these studies in mere stage of technique and evaluation. In this technical

field, not only more researchers should be engaged in the evaluation of physical properties of organic molecules, but also they should have more clearly defined image of the device and should continue to study with more challenging spirit.

Then, for the purpose of creating single molecular electronics and electronic system based on it, which type of active electronic functions should we seek in the single molecule? As one of the replies to such question, our study group is aiming to achieve organic electronic device of the next generation in information and communication field. We are trying to take advantage of the characteristics of single organic material and are seeking the features, which the conventional Si type inorganic material could not attain, and we are also trying to develop single electron tunneling (SET) device using the organic molecule. The SET device itself has a structure with small particles formed in electrical tunneling barrier as Coulomb Island, and various applications have been conceived including memory device, high-speed low-power switching device, high-sensitive electrometer device, etc. [5] [6]. With the progress of ultra-fine lithographic technique to produce semiconductor devices, the development of new devices is now in progress based on quantum effect, and this is one of the important research fields for inorganic semiconductor materials. In addition to such features, based on the use of organic molecule, it is now possible to add the features of organic molecule such as ultra-fine property, photo-responsibility, self-organizing ability, flexibility, etc. This makes it possible to attain new functions while taking advantage of the conventional type SET device. In the present report, we take special notice on ultra-fine property and photo-responsibility of organic molecule and introduce the results of the study to the device using single organic molecule as Coulomb Island for SET device. Further, we will discuss the possibility of attaining organic SET device.

2 Development of Single Electron Tunneling Device using Organic Molecule as Coulomb Island

[7]-[10]

As already described, the study is now being made on molecular electronics with aims to provide electronic functions and optical functions of a single molecule by using organic molecule and to apply the functions to electronic-optical device. Above all, attention is focused on the functions of single molecule, and the study of single electron tunneling (SET) device in the single molecular electronics for application of electronic device is now one of the important research fields together with the study on the development to system integration and on the application of various types of devices.

2.1 Single Electron Tunneling Device

Single electron tunneling device is defined as a device, which can control single electron transfer between electrodes using Coulomb blockade phenomenon. As shown in Fig. 1 (a), when a small Coulomb Island is placed between two electrodes of a device for tunnel conduction, if electron is stay in the Coulomb Island, transfer of tunneling electron to the Coulomb Island is restricted by electron charging energy $e^2/2C$ of the Coulomb Island. This leads to remarkable discreteness in electric current transport and to functional features different from those of general electronic device (Fig. 1 (b)). However, in order to obtain such electrical characteristics, it is necessary to have the electron charge energy $e^2/2C$ to be at a level higher than thermal energy kT . As a result, the size of small particles functioning as the Coulomb Island must be set to the size of several nm, i.e. not more than 10^{-18} F as electrical capacity [11] [12].

In this respect, we produced one of organic molecular devices by using a nano-meter-size organic single molecule as the small Coulomb Island. When organic molecule is used as the Coulomb Island, very small Coulomb Island can be easily produced. If a dye molecule is used, it is possible to add

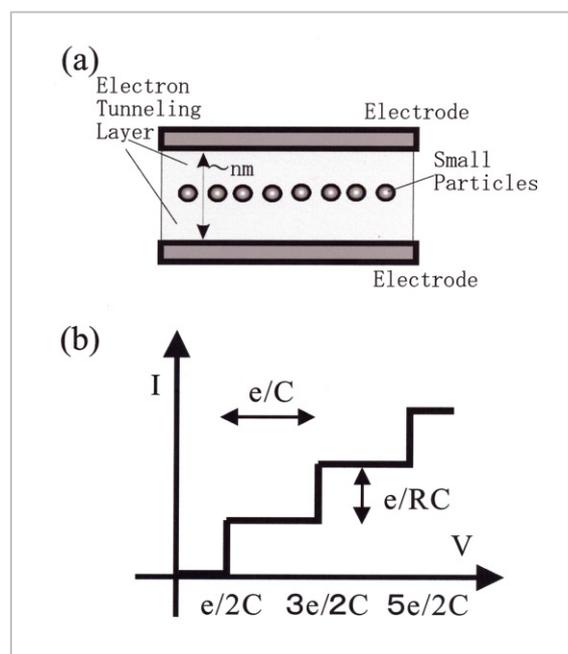


Fig. 1 Structure of single electron tunneling device (a) and I-V characteristics (b)

electronic-optical function of the molecule itself to SET characteristics, and a new function may be provided, which has not been found in the devices using inorganic material.

2.2 Fabrication of Organic Single Electron Tunneling Device

Here, we will describe fabrication organic single electron tunneling device having dye molecule as Coulomb Island using organic electron tunneling barrier. Also, electrical property of this device will be discussed. Organic molecule is not necessarily used for the electron tunneling layer. However, organic electron tunneling layer was adopted because of the possible development to future application of device such as photo-isomerization switching, flexibility, etc. In single electron tunneling device of photo-switch type as to be described later, inorganic electron tunneling layer was adopted by giving consideration on the improvement of production yield, high-temperature operation, and development to memory device.

2.2.1 Experiment [8]

As the molecule used for the fabrication of the device, dendrite molecule, for which special notice has been given in the field of

organic synthesis in recent years, was used as Coulomb Island, and polyimide (PI) LB film [14] was used as electron tunneling barrier because it is known to provide satisfactory functions as electron tunneling barrier [13] and it can control single molecular film thickness [0.4 nm] by LB method. Dendrite molecule used as the Coulomb Island has spherical shell molecular structure with rhodamine dye molecule enclosed by CH group with electrically insulating property. For this reason, it is possible to electrically isolate the dye molecule at the center from outside. That is, this molecule has the possibility to become organic single molecular single electron tunneling device by making direct electric access to the molecule when the device is prepared as a single organic molecule. Also, it is known that it is a molecule, which is difficult to be associated because the molecule itself is covered with hydrophobic CH group [15]. The structure of the organic molecule used in the present study is shown in Fig. 2.

Using the organic molecule as described above, a single electron tunneling device was prepared, which has the device structure as shown in Fig. 3 and has rhodamine dye dendrite molecule as Coulomb Island layer. The device was prepared on a glass substrate. Upper and lower Au electrodes were produced by vacuum evaporation method, and electron tunneling PI layer was produced by LB method. In the Coulomb Island layer, PI LB film mixed with rhodamine dendrite molecule was accumulated in mono-layer. Molecular mixing ratio was set to: PI:Rh-G2 = 500:1 so that Rh-G2 molecule provides functions as the Coulomb Island and that molecules are not associated with each other. From the experiment on molecule occupying area, it is known that about 1000 Rh-G2 molecules are present within an area of about 1 square μm in molecular ratio, and it is considered that the molecules effectively work as the Coulomb Island. As a result, effective area of this device approximately 50×100 square μm . The electrical resistance of this device is several hundreds of $\text{M}\Omega$ to several tens of $\text{G}\Omega$.

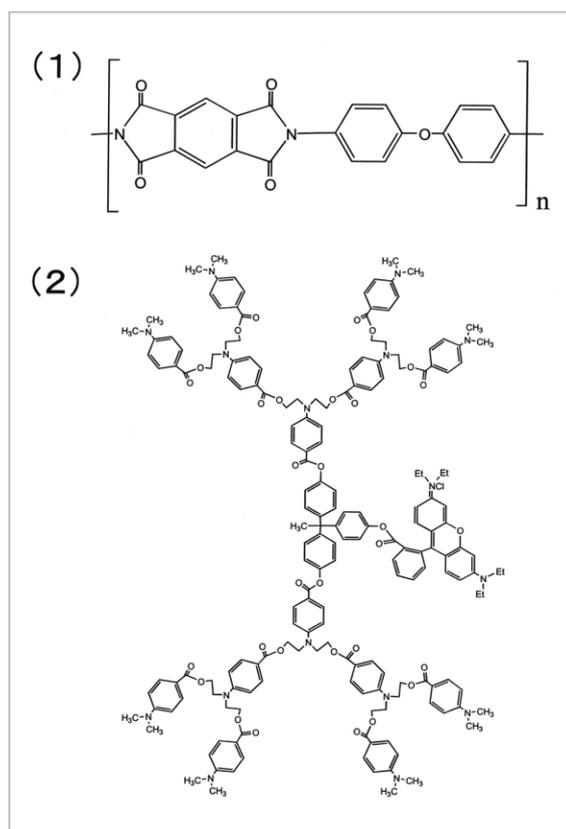


Fig.2 Structure of molecules
 (1) Polyimide (PI)
 (2) Rhodamine dendrite molecule (Rh-G2)

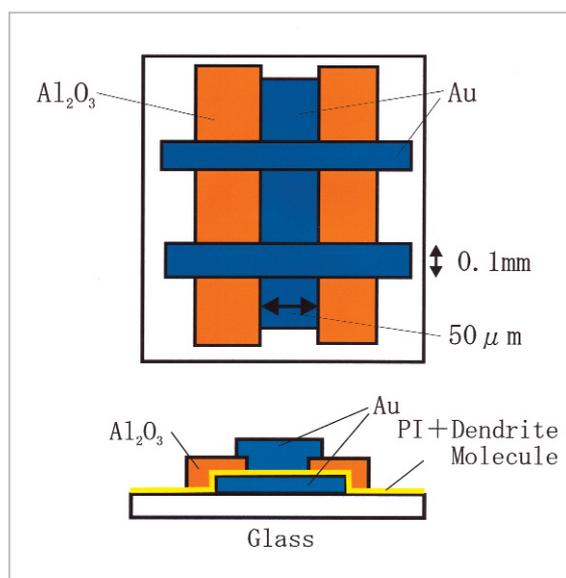


Fig.3 Structure of organic single electron tunneling device

Two-terminal method was used for measurement, and measurement was performed in a cryostat at a constant temperature by applying step voltage.

2.2.2 Electrical Property of Single Electron Tunneling Device

Figure 4 shows I-V characteristics of Au/PI25/PI+Rh-G2/PI30/Au device at the temperature of 5.2 K. Voltage step of equal spacing, which is characteristic of SET property, is clearly seen. Voltage width based on SET phenomenon is a constant voltage width of e/C , here C is the electrical capacity between the Coulomb Island and electrodes [11] [12]. As seen in the figure, the voltage step width is about 100 mV in both 0-th order and 1st order, and this clearly indicates that SET property appeared. To elucidate this, finite difference of the data in the figure was calculated and plotted. The results are shown in Fig. 4 (b). Peaks of dV/dI are seen with equal spacing at 50, 150 and 250 mV in both positive and negative voltages, and the property of the SET device can be clearly found.

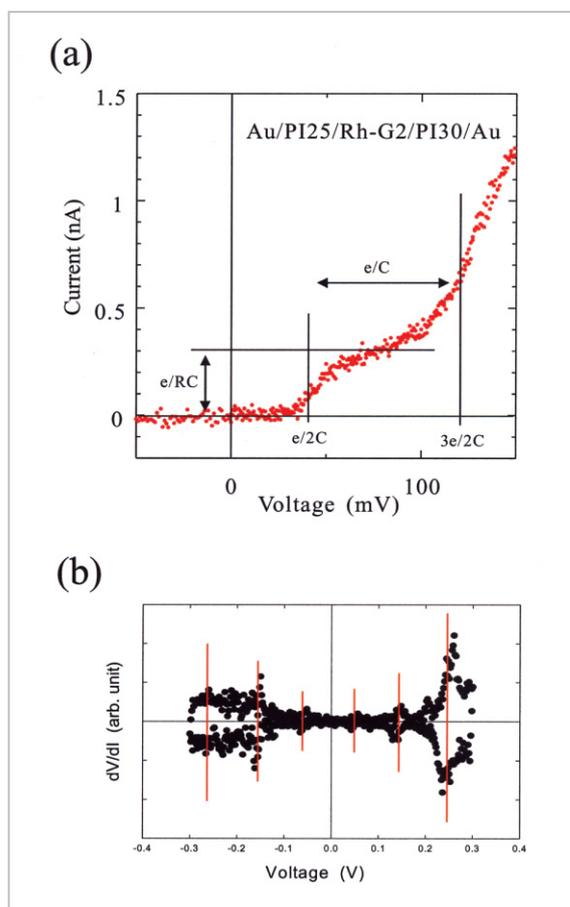


Fig.4 (a) I-V characteristics of Au/PI/Rh-G2/PI/Au structure (b) dV/dI -V characteristics of Au/PI/Rh-G2/PI/Au structure

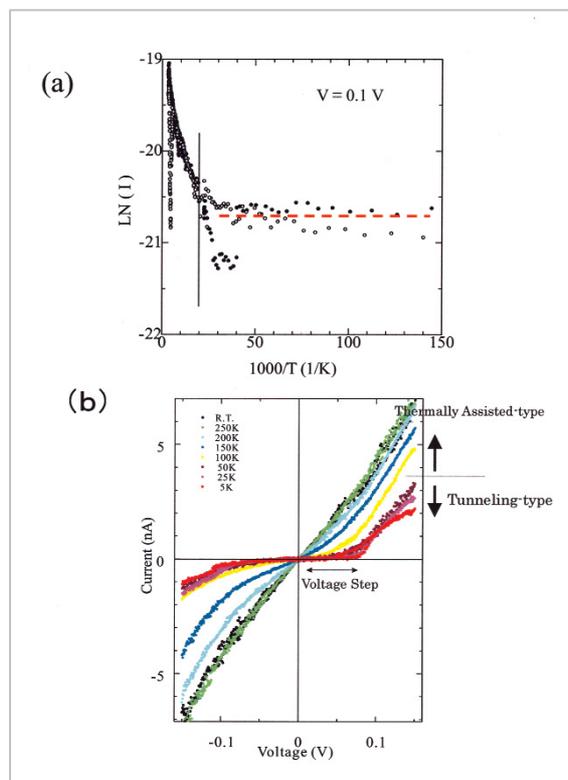


Fig.5 Temperature dependence of electrical properties of organic single electron tunneling device (a) I - $1/T$ plot (Arrhenius' plot) (b) Temperature dependence of I-V characteristics

Next, temperature property of the device was measured to identify tunnel conduction of the device. The results are summarized in Fig. 5. From the figure, it is clearly seen that electric current decreased with the decrease of temperature from room temperature to 50 K, and this indicates that it is electrical conduction property of thermal activation type. However, when temperature is further decreased, almost no change of electric current is observed at the temperature of 50 K or lower, and it is evident that it indicates electrical conduction of tunnel conduction type. When thermal activation energy at the temperature 50 K or higher is calculated, it is a very small value of 30 meV or lower. This suggests that both conduction components of thermal activation type and tunnel type are exist in this device, and that electric conduction component of thermal activation type, which was prevalent at room temperature, was gradually reduced and electric conduction of

tunnel conduction type became dominant, and this would have ultimately lead to SET property as shown in Fig. 4. To confirm this, I-V characteristics were measured at each constant temperature. The results are shown in Fig. 5 (b). It is evident that step property of the voltage, i.e. single electron tunneling property, is observed on low temperature side lower than the temperature of 50 K, which indicates electric conduction of tunnel conduction type. The voltage step width is not different from the value observed at the temperature of 5.2 K, and the same SET property with no change in temperature can be obtained at each temperature. Thus, it was found that single electron tunneling property could be obtained at temperature condition where tunnel conduction property is attained in the device using Rh-G2 dendrite molecule. In general, for the purpose of accurately attaining SET phenomenon, the energy increase of $e^2/2C$ associated with electron tunneling must be equal to or more than the thermal energy kT [11] [12]. In the SET device as generally prepared, this condition is often the temperature condition for device operation. However, in the device presently prepared, voltage step due to SET property is about 50 meV, and this suggests that it is possible to operate at room temperature if the temperature range to attain the tunnel conduction property can be increased.

As described above, it was found that the device using organic single molecule, Rh-G2 dendrite molecule, as Coulomb Island can provide functions as SET device, and that the device potential is so high as to achieve operation at room temperature.

2.2.3 Organic Single Molecular Coulomb Island [8] [10]

It is evident that the device thus prepared has the SET property as shown in Fig. 4 at low temperature. Evaluation is now made briefly as to whether Rh-G2 molecule introduced as the Coulomb Island actually provides the functions as the Coulomb Island. One approach to such evaluation is to estimate the size of the material, which provides the func-

tion as the Coulomb Island. Detailed study has also been performed [16], while verification is made here by simple approximation. As already described, voltage step width as seen in the device property is in the size of e/C . On the other hand, electric capacity C of a conductive material with a radius of "r" positioned at a distance "d" on a flat electrode can be expressed as given below under the condition of $r \ll d$:

$$C = \frac{4\pi\epsilon_r\epsilon_0}{(1/r - 1/2d)}$$

Therefore, the radius "r" of the Coulomb Island is given as:

if it is assumed that voltage step width actually observed is ΔV .

$$r = 1 / \left(\frac{4\pi\epsilon_r\epsilon_0}{e} \Delta V + \frac{1}{2d} \right)$$

Based on this equation, it is assumed that Rh-G2 molecule regarded as the Coulomb Island is considered as a conductive material in spherical shape. By substituting the equation with the above observed values (i.e. $\Delta V = 100$ mV, $\epsilon_r = 3$ (polyimide), and $d = 10$ nm (PI 25 layers)), the size of r of the Coulomb Island is given as: $r = 3.8$ nm. Actual size of Rh-G2 molecule is considered as about 1 - 2 nm (radius) from simulation and from the experiment on LB F-A technique. Thus, the size is somewhat smaller. This may be attributable to the facts that calculation was based on postulation and Rh-G2 molecule was regarded as conductive material, and also, that effective film thickness of polyimide LB film, providing the function as electron tunneling layer, was decreased physically and electronically [17] [18]. Thus, the postulation for the calculation cannot be accurately established. If the decrement of the effective PI electron tunneling layer is estimated from the size of Rh-G2 molecule, it appears that the films are decreased by about 15 - 20 layers. This value may be regarded as adequate because the decrease of film thickness of the same extent is seen in the results of the study.

As described above, there may remain

some uncertainties about the decrease of PI film thickness, but it is considered that Rh-G2 molecule is functioning as the Coulomb Island in this device. Also, although it is not shown in the figure, the property as shown in Fig. 4 is not seen at all in the device where Rh-G2 molecule is not introduced, and it provides the function as a mere tunnel conduction device.

3 Optical Gate Effects of Single Electron Tunneling Device

As already described, when organic molecule - in particular, dye molecule - is used as the Coulomb Island for SET device, it is possible to add electronic-optical functions of the organic single molecule to the functions of SET device. One of such additions is the addition of molecular function based on light irradiation. For instance, there are possibilities that the structure of dye molecule itself is changed due to light irradiation (a), or that single electron tunneling phenomenon is changed due to the induced electrons from dye molecule (b), or that electron tunneling barrier is changed due to the induced charge, and electron tunneling probability is changed (c) (Fig. 6). If these phenomena are applied, by using the organic molecule as the Coulomb Island, new optical functions can be added to SET device, or single electron control can be achieved by optical gate effect instead of electronic gate, which is difficult to make access to single molecule. In addition, through the access to multiple molecules by light, the ways to new architecture may be opened toward cell automaton, parallel processing, or network processing.

3.1 Light Irradiation Property of Organic Electron Tunneling Layer SET Device [8]

A device to measure light irradiation property was prepared by the same procedure used in the preparation of the device as described above by using ITO electrode as a lower electrode. The light was irradiated through ITO electrode from the side of glass substrate at a

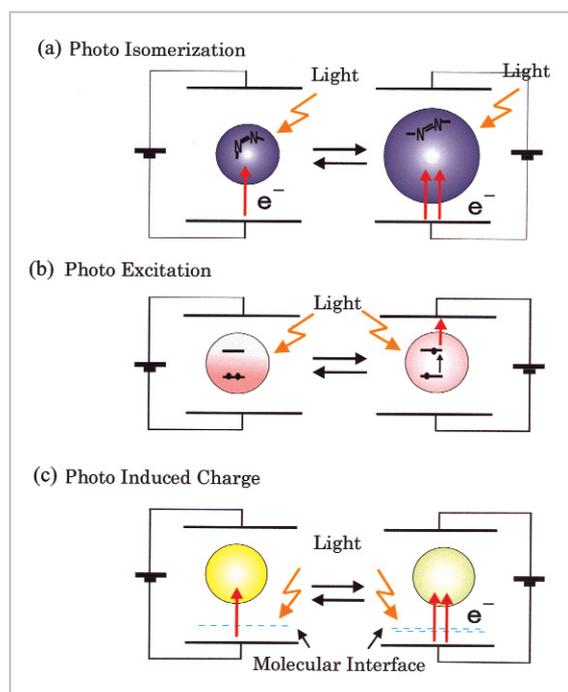


Fig.6 Schematic drawing of optical gate type SET device

constant temperature, and electric property during light irradiation was determined.

White light was irradiated at first to the organic single electron tunneling device having the organic electron tunneling layer as described above, and the change of the property was carefully observed. The results are shown in Fig. 7. By irradiating white light, the change of the single electron tunneling property could be observed, and it was confirmed that the initial property was restored when it was brought to dark place. As seen in the figure, electric current value was decreased due to light irradiation, and tunnel conduction property was changed. However, position and width of voltage step, which are the features of SET property, were not changed. This suggests that the change due to light irradiation was not caused by the change of Rh-G2 molecule, working as the Coulomb Island, but it was caused by the change of PI electron tunneling barrier. For instance, it is considered that such optical property is developed by the change of electric charge property induced in PI molecular film working as electron tunneling layer by the change of charge transfer [14] at metal/PI interface due to light

irradiation. The details must be verified further, but this may suggest the possibility that this change in potential can be used for the control of single electron tunneling property because potential is changed due to charge transfer at the interface.

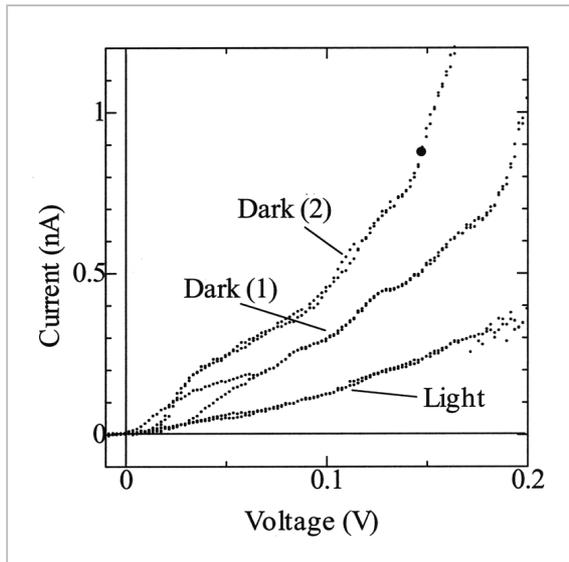


Fig.7 Changes of organic SET device property due to light irradiation
- Organic electron tunneling layer organic SET device -

3.2 Light Irradiation Property of Inorganic Electron Tunneling Layer SET Device [20]

When single electron tunneling device is prepared by using organic molecule as Coulomb Island, a part of new properties such as flexibility and controllability of property due to structural isomerization may be sacrificed, but there is no need to use organic molecule as the electron tunneling layer. Instead, by using inorganic material, it is possible to receive benefit of Si technology obtained so far such as material stability, controllability, etc. Thus, device property by organic dye molecule used as Coulomb Island can be properly evaluated. In this respect, with the purpose of clearly identifying the change of property of single electron tunneling device due to light irradiation using organic molecule as Coulomb Island, SiO₂ was used as the electron tunneling layer, and evaluation was made on

the stable device property. A highly doped Si substrate with flat surface was used as lower electrode in order to prepare the upper electron tunneling layer with very thin thickness and with high controllability. A SiO₂/organic molecule/SiO₂ layer was deposited on it by vacuum evaporation. Finally, a gold electrode was deposited and this was used as upper electrode, and the device was prepared [20] [21] (Fig. 8 (a)). The light was irradiated through the upper semi-transparent gold electrode, and monochromatic light to be absorbed by dye molecule used as Coulomb Island was irradiated. In a cryostat and at a constant temperature, the electrical property was measured. Figure 8 (b) shows the results of measurement using porphyrin (POR) type molecule as organic dye molecule of Coulomb Island. By irradiating the light with absorption wavelength of the dye molecule used as Coulomb Island, the change of single electron tunneling property was observed. It was confirmed that initial property was restored when it was brought back to dark condition. Unlike to light irradiation property as described in the preceding section, the change of position and width of voltage step, which is characteristic to the single electron tunneling property due to light irradiation, are seen in the figure. This means that, unlike the change in the values of electron tunneling current caused by the changes of electrical tunneling barrier as described in the preceding section, the conductive condition of the single electron tunneling has changed. Specifically, by projecting the light, single electron blockade phenomenon of organic SET device is controlled and gate control of SET device is carried out based on the light. Therefore, by applying a certain voltage and by repeating light irradiation, single electron tunneling condition can be changed between before and after the light irradiation. By the gate effect of the light of SET, on/off switching of electric current value flowing in SET device can be seen. The results are shown in Fig. 8 (c). As described above, switching phenomenon caused by optical gate was observed in organic single elec-

tron tunneling device using organic molecule as the Coulomb Island. The details of the mechanism are to be elucidated further. Although it is not shown here, changes may have occurred in the single electron tunneling device due to the charging effect into the tunnel barrier [22]. The study is now being made on the development of optical gate type organic single electron tunneling device for operating the organic Coulomb Island in more direct manner.

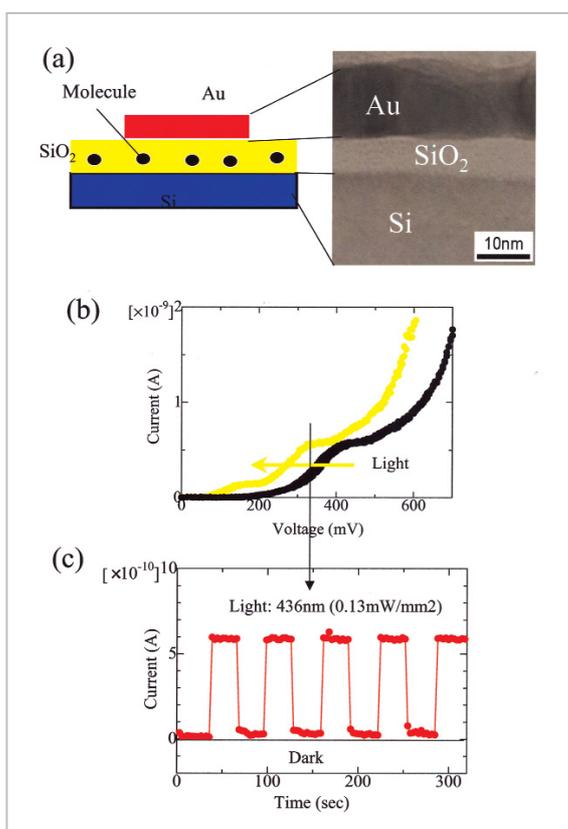


Fig.8 Changes of organic SET device property due to light irradiation
 — Inorganic electron tunneling layer organic SET device —
 (a) Structure of inorganic electron tunneling layer organic SET device
 (b) SET property under light irradiation
 (c) Optical gate switching property

4 Engagement Toward Single Organic Molecular Electronics

Up to the preceding chapter, description has been given on the fabrication of organic single electron tunneling device based on the

property of single organic molecule and electronic-optical property of such device. The property is based on organic single molecule working as Coulomb Island, and it is important for constructing organic single molecular electronics. However, in attempting to systematize and to integrate the organic single molecular electronic device further, it is also important to have the technique to gain direct access electronically to each individual organic molecule and to directly evaluate electronic-optical property and to effectively connect the organic electronic device. Here, we briefly introduce the preparation of ultra-fine nano-gap electrode as currently performed in our laboratory and also describe the preparation of organic conductive nano-wire.

4.1 Preparation of Ultra-fine Nano-Gap Electrode [23]

The measurement of electrical property of single organic molecule isolated on an insulated substrate is very important for the evaluation of electronic property of single organic molecule and also for the evaluation of basic device property directed toward integration of molecular device in future. To have access electronically to single organic molecule, an approach by using probe such as STM is commonly practiced in the current technique. However, when we consider the difficulty to have approach to single molecule isolated on an insulated substrate or when we think about the shifting to circuit network directed to the preparation of device in future such as the arrangement of gate electrode, it would be necessary to prepare ultra-fine nano-gap electrode with gap length of several nm or shorter, which enables direct access to single molecule of about several nm in size, and also it would be necessary to evaluate electrical property of single molecule. The preparation of such ultra-fine nano-gap is difficult to accomplish by conventional lithographic method using electron beam as practiced in the past, and various types of methods are now being investigated [24] [25]. Above all, in order to prepare a nano-gap electrode on nano-

meter level, which is in steady state and shows good reproducibility, we successfully developed a new process using the focusing ion beam (FIB) system and prepared nano-gap Au electrode of several nm in width on a SiO₂ insulated substrate [23]. Although the details of this process are not given here, SEM image of Au nano-gap electrode prepared on SiO₂ substrate is shown in Fig. 9. A satisfactory Au nano-gap electrode of about 5 nm in gap length has been prepared, and it showed high reproducibility. Currently, dye organic molecule as described above and other functional molecules and materials are introduced between the nano-gap electrodes thus prepared, and evaluation is being made on electrical property.

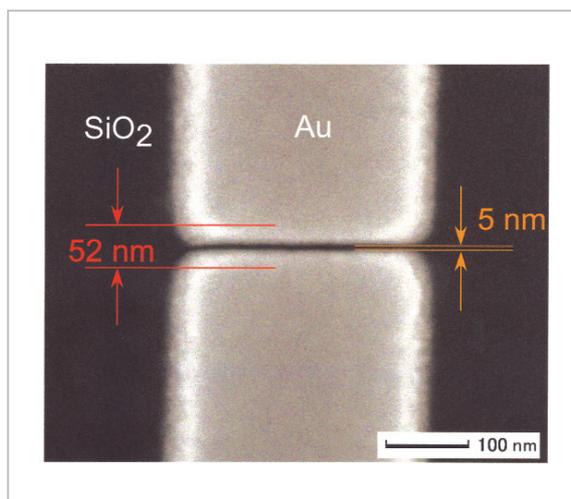


Fig.9 SEM image of the prepared Au nano-gap electrode

4.2 Preparation on Nano Size Organic Conductive Wire [26]

In the integration of organic electronic device, it is an important problem how to achieve the wiring between devices. As one of the approaches, efforts are now being made on the preparation of the organic conductive wire and to turn the wire to nano size. For the wiring between organic molecular devices, it is desirable to construct self-organizing and conductive nano-wire between the devices by the wiring using top-down type metal. For this purpose, studies are currently performed

on various types of molecular wires with central core in the conductive macromolecular material [18]. In our laboratory, an attempt is now made to prepare nano-wire of conductive molecular crystal by electrochemical method. Under the conditions for preparation, we have successfully prepared conductive molecular nano-wire of about several tens of nm as shown in Fig. 10 [26]. At present, we are working not only on the evaluation of conductivity in nano region, on the preparation of nano-wire in smaller size and further, on wiring between devices, but also on embedding of electric functions into nano-wire itself.

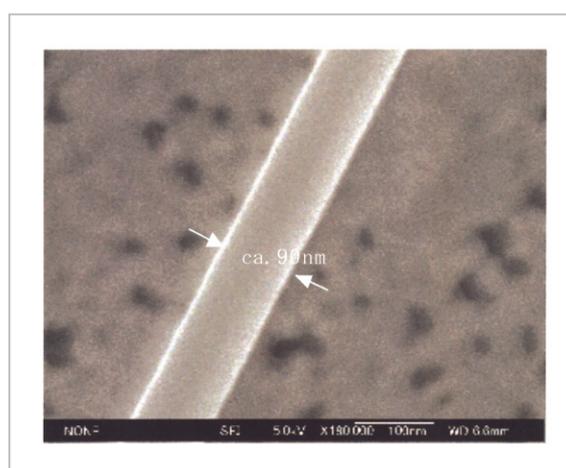


Fig.10 SEM image of the prepared organic conductive crystal nano-wire

5 Summary

The attempts to use organic molecule as active material have shown rapid progress in the past several years. On the background of such progress, we can see practical use of liquid crystal display originated from the discovery of conductive molecules and successful preparation of EL device, which is currently shifted to practical use and showing enormous achievement in the development. When we look to future aspect of the electronics, molecular electronics using organic molecule as active device has the possibility to develop as the electronics of the next generation with high achievability because it is not only to replace the current stage of electronics but

also because molecular device is cited in the road map of semiconductor device [27]. Further, new molecular materials such as nanotube, fullerene, dendrimer, etc. have been detected, and proposal and trial production are successively made on new functional devices such as molecular photonic device and bio-functional molecular device. It will be developed to molecular electronics, which would include not only the technique to control molecules and to draw out the properties specific to molecule but also the quantum effect based on ultra-fine molecular size. We can see several pioneer studies, including the studies on

single molecular operation, self-organizing, molecular wire, single molecular rectifying device, single molecular light emitting device, molecular FET, molecular transistor, etc. The SET device using organic single molecule as introduced in the present report may be one of such subjects of the studies. It is expected that the molecular device thus prepared will be systematized together with the results of other studies and would act as a part of the organic molecular devices, which would play an important role in the field of information and communication in future.

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