Room-temperature observation of a Coulomb blockade phenomenon in aluminum nanodots fabricated by an electrochemical process

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An aluminum nanodot was self-organized between two electrodes using the anodization process of an aluminum microelectrode of 3 μm in width. The authors observed a clear Coulomb staircase with a very large Coulomb energy of about 2 eV at room temperature. This very large Coulomb energy is attributed to the device structure which depends strongly on the aluminum nanodot formation mechanism. The authors' results indicate that a single electron transistor operating at room temperature can be fabricated at an appropriate position using both bottom-up and top-down processes. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2475419]

A technology for fabricating nanostructures has been extensively studied in order to develop electronic or photonic devices based on new types of mechanisms such as quantum effects. There are generally two complementary approaches to fabrication of nanostructures. One is a top-down process as typified by a lithography technique widely used in large scale integration technology and the other is a bottom-up process such as a self-organization process. In the case of the conventional lithography technique, it is not easy to fabricate nanostructures although it is suitable for controlling their positions. In contrast, a self-organization process does not control positions of nanostructures though it can easily produce nanostructures. Therefore, it is important to develop a hybrid technique of these complementary techniques. Matsumoto *et al.* fabricated a single electron transistor (SET) of a carbon nanotube (CNT), grown across a gap of several micrometers in an iron electrode using a lithography technique. A Coulomb energy of 400 meV was observed, which was much larger than a thermal energy of 26 meV at a room temperature.[1] However, the control of the electrical properties of CNT has not been established, and the development of a new process to introduce self-organization phenomena is required.

It is well known that an anodization process of aluminum easily forms porous anodic alumina with self-ordered nanohole arrays.[2] A Coulomb blockade phenomenon was observed at room temperature in nanodots or nanowires of cadmium sulfide or zinc selenide deposited in the holes of the porous anodic alumina although the positions of the nanodots or nanowires were not controlled.[3] This letter exhibits the potential of porous anodic alumina as a template for a nanodevice. Here, we report on a method of fabricating nanodots controlling their positions using both the anodic porous alumina formation process and a photolithography technique, and the electron transport characteristics of this structure at room temperature were investigated.

Aluminum nanodots are self-organized after anodizing an aluminum film on a substrate.[4,5] In this study, the position of aluminum nanodots was controlled by anodizing the part of an aluminum microelectrode by means of a photolithography technique as shown in Fig. 1. Figure 1 shows a schematic of a microelectrode fabricated by a photolithography technique before anodization. A cross-sectional view in Fig. 1 illustrates a porous anodic alumina layer and an aluminum nanodot formed by partial anodization of the aluminum microelectrode.

A *p*-type silicon (100) wafer was used as a substrate. A 500-nm-thick aluminum film was evaporated on a 100-nm-thick silicon dioxide layer formed on the silicon substrate surface. The aluminum film was etched using the photolithography technique to form an aluminum microelectrode of 3 μm in width. The microelectrode was covered with a 200-nm-thick silicon oxide (SiO_x) layer deposited by the resistance heating method, except the layer was 3 μm in length at the midpoint of the microelectrode to control the anodized area. Finally, the uncovered area of the electrode was anodized in a sulfuric acid solution of 1.7% at an anodic potential of 25 V using platinum as a counterelectrode.

In order to stop the anodization of the microelectrode at an appropriate time to fabricate the SET structure, we moni-

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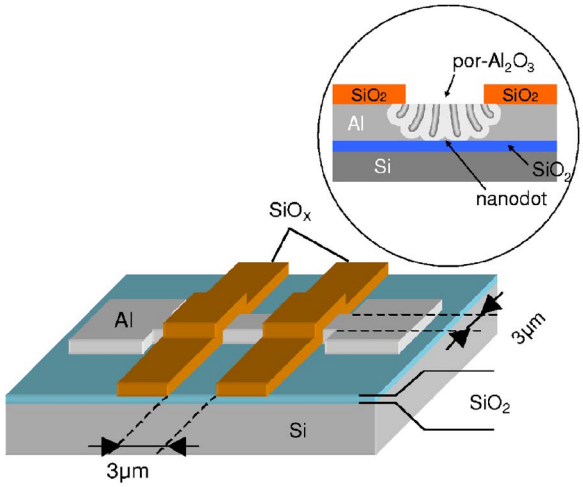

FIG. 1. (Color online) Schematic of a microelectrode fabricated by a photolithography technique before anodization and a cross-sectional view of the partially anodized microelectrode.

tored the potential of the microelectrode. As shown in Fig. 1, the microelectrode had two pads for applying and monitoring the potential of the microelectrode. One of the pads was used to apply an anodic potential to the microelectrode, while the other pad was used to monitor its potential. When the microelectrode is broken by anodization, the monitored potential decreases even though it is equal to the applied anodic potential at the initial stage of anodization. The potential drop indicates the time of the formation of the nanostructure. Thus, the anodization time can be controlled by the difference between the monitored potential of the microelectrode and the applied anodic potential. The difference of the potentials was set at 0.1 V.

Figure 2 shows the *I-V* characteristics of the anodized microelectrode, which were measured at room temperature. A clear staircase current was observed at source-drain voltages of 2 and 6 V, which indicates a Coulomb blockade phenomenon. Its Coulomb energy is about 2 eV, which is the largest value in comparison to the values found in our literature survey. A total capacitance of this device is estimated at about 0.04 aF, which corresponds approximately to a capacitance of parallel plate electrodes with an area of $2.3 \times 2.3\ \mathrm{nm}^2$ and a gap of 10 nm. The small capacitance is

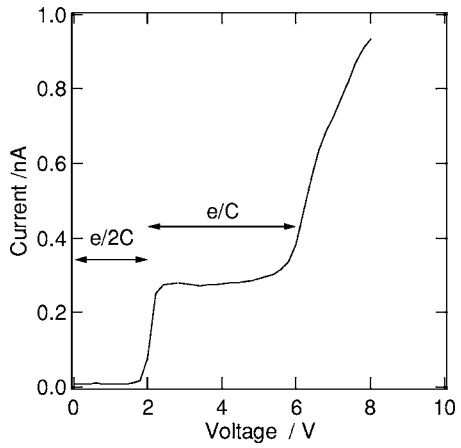

FIG. 2. *I-V* characteristics for a SET structure fabricated by both anodization process and a lithography technique.

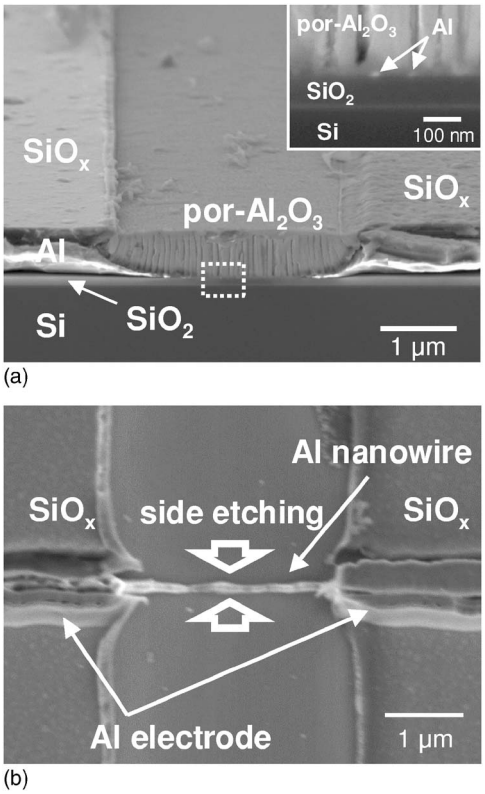

FIG. 3. FE-SEM images for a (a) cross-sectional view of a porous anodic alumina layer formed by partially anodizing an aluminum film and (b) top view of an anodized microelectrode before it was broken.

attributed to the structure of electrodes and nanodots formed by anodization.

Figure 3(a) shows a cross-sectional field-emission scanning electron microscope (FE-SEM) image of a porous anodic alumina layer formed by partially anodizing an aluminum film, where the aluminum film was anodized in an oxalic acid solution at 40 V for easy observation of the structure of a porous alumina layer because the pore size of porous anodic alumina is proportional to an anodic potential.[6] A porous anodic alumina layer with bended pores grows around the edges of the SiO_x mask window although the straight pores grow out perpendicularly around the center of the SiO_x mask window. Thus, the vertical growth rate of porous alumina around the center is larger than that around the edge. The partially anodized electrode is consequently very thin at the midpoint where the nanodots form as illustrated in the cross-sectional view of the structure shown in Fig. 1. The insertion in Fig. 3(a), represented by a box of white dashed line, is an enlargement of the interface between a porous anodic alumina layer and a silicon dioxide layer. Aluminum nanodots are clearly seen in the insertion in Fig. 3(a). The structure depicted in Fig. 1 was formed in the partially anodized aluminum microelectrode. In addition, it is important to fabricate narrow and thin source and drain electrodes for observing Coulomb staircases because narrowing and thinning of electrodes decrease the capacitance of the devices. The side etching of the microelectrode takes place simultaneously to vertical etching because the side of the microelectrode is not covered with the SiO_x film as shown in Fig. 1. It forms a nanowire leading to a decrease in capacitance of the devices. Figure 3(b) shows a FE-SEM image for a microelectrode before the microelectrode was broken,

where the porous anodic alumina layer was removed with a mixture of chromic acid and phosphoric acid. We can see a self-organized nanowire of about 100 nm in width in Fig. 3(b). Figures 3(a) and 3(b) indicate that the anodization process causes simultaneous thinning and narrowing of the center of the aluminum microelectrode. The nanodots form just when the microelectrode is broken. The thinning and narrowing of the microelectrode decrease the capacitance of this device and ensure that nanodots form under the mask window. As a result, a SET structure with very high Coulomb energy can be fabricated at an appropriate position even when the structure is so small that it cannot be manufactured solely through the photolithography technique.

In summary, we fabricated a SET structure at an appropriate position through both anodization of the aluminum microelectrode and photolithography techniques. Then, the *I-V* characteristic of the device at room temperature was investigated. We observed a clear Coulomb staircase with a Coulomb energy of about 2 eV. Our results indicate that a novel technique which involves a hybrid approach of self-organization phenomenon and lithography techniques is capable of producing small nanodevices which cannot be fabricated solely through conventional lithography techniques. Our technique can be applied to another valve metal such as titanium and gives new selections of materials and methods of nanodevices leading to production of nanodevices with new functions.

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