# Nanoparticle Single-Electron Transistor with Metal Bridged Top-Gate Electrodes

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## 1. Introduction

A combination of bottom-up processes with top-down processes is one of the candidates to fabricate nanodevices with precise structures in a subnanometer scale. Single-electron transistors (SETs) have been studied extensively, because SETs are nanodevices that can be applied to Boolean logic with high device density and low power consumption at high speed[1]. Au nanoparticles have a conductive Au core-insulative shell structure in which both the diameter of the Au core and the tunneling resistance of the insulative shell are controlled by chemical syntheses and ligand molecules, respectively, and are key components fabricated by the bottom-up processes for Coulomb islands in SETs[2,3]. For a fabrication of integrated nanogap electrodes on a sample simultaneously, we have established a process of integrated nanogap electrodes by a combination of the top-down processes of electron beam lithography (EBL) technique and the bottom-up processes of electroless Au plating with a 41% process yield using a common medical liquid[4]. Combining the Au nanoparticles and the nanogap electrodes, we have demonstrated SET operation at 80 K[5].

In an operation of SETs, a charging energy in Coulomb islands has to be much larger than a thermal energy of  $k_BT$ . The charging energy is inversely proportional to a capacitance of the Coulomb islands which is proportional to their diameter. Therefore, for a realization of SET operations at room temperature, the size of the Coulomb islands has to be as small as  $\sim 2$  nm. On the contrary, when the size of the Coulomb islands reduces, a gate capacitance is also reduces. Small gate capacitances are not preferable since a gate voltage for control on/off conditions increases. Therefore, improvements of gate electrode structures are required for increasing the gate capacitance. One solution for increasing is to surround Coulomb islands by a gate electrode where electric fluxes from the Coulomb islands terminate to gate electrodes. Here, we fabricate a metal bridged top-gate electrode by using two consecutive electron beam lithographies on Au nanogap electrodes. With these Au nanogap electrodes and Au nanoparticles we demonstrate SET operations and discuss an improvement of gate capacitances.

## 2. Experimental

We used EBL and a lift-off process to fabricate the Ti /Au electrode patterns of source, drain, and side gate electrodes (upper and lower electrodes is denoted as gate 1 and gate 2, respectively) with a gap of 30 nm between the source and drain electrodes on a SiO<sub>2</sub> (300 nm)/Si substrate (Fig. 1(a)). Fabrication of metal bridge structure consists of two consecutive electron beam lithographies. The sacrificial layer of 200 nm thick polymethylmethacrylate (PMMA, 495K, Microchem) was spun on and cross-linked between



Fig. 1 (a)-(d) Schematic illustrations in fabrication processes of a metal bridged top-gate electrode. (e) SEM image of a typical nanogap electrode corresponds to (a). (f) SEM image of typical cross-linked PMMA corresponds to (b). (g) and (h) SEM images of typical metal bridged top-gate electrode at top-view and oblique view, respectively.

source and drain electrodes. High doses of electrons were bombarded to the designated areas, which will remain, with the rest dissolved away in acetone (Fig. 1(b)). It notes that the effective thickness of this PMMA sacrificial layer after being cross-linked was reduced to a half of the initial thickness, that is, approximately 100 nm thick[6]. Then, PMMA layer with 460 nm thick was spun on the sacrificial layer and an electrode pattern was drawn across the sacrificial layer between the gate 1 and 2 electrodes with low doses. Ti/Au was evaporated on the sacrificial layer, then the sample was lifted-off by acetone to remove unwanted metals (Fig. 1 (c)). After that, the cross-linked PMMA sacrificial layer was removed using O<sub>2</sub> plasma ashing. With these processes, a bridge structure across the nanogap region between the gate 1 and 2 electrodes was fabricated (Fig. 1(d)).

The sample with the metal bridged top-gated electrode was immersed in a 5 mM solution of C8S in ethanol for 24 h to form C8S self-assembled monolayers (SAMs) and then rinsed with ethanol twice. Subsequently, the sample was immersed in a 500 mM solution of decanedithiol [HS(CH<sub>2</sub>)<sub>10</sub>SH, C10S2] in ethanol for 24 h to partially substitute C8S by C10S2 then rinsed with ethanol twice[7]. After the substitution, the sample was immersed in a solution of C8S-Au nanoparticles (diameter 5.2±0.5 nm) in toluene for 15 h. Some C8S molecules surrounding Au cores were expected to be substituted with C10S2 molecules on the Au nanogap electrodes; therefore, C8S-Au nanoparticles were expected to be anchored by C10S2 molecules. Electrical measurements were carried out using a semiconductor parameter analyzer (Agilent B1500) under a vacuum ( $\sim 10^{-5}$  Pa) at 9 K by a helium refrigerator.

#### 3. Results and Discussion

Figures 1 (g) and (h) are scanning electron micrograph (SEM) images of a metal bridged top-gate electrode across the drain-source electrodes. The metal bridge structure was fabricated between the gate 1 and 2 electrodes.

Figure 2 shows that experimental differential conductance  $dI_d/dV_d$  plot in the  $V_g$ - $V_d$  plane, which is so-called stability diagram where the differential conductivity smaller than 30 pS corresponds to purple in Fig. 2. Clear Coulomb diamonds are observed. The theoretical solid lines in Fig. 2 represent the conductance peak lines calculated from a capacitance between the drain and the Au core  $C_1=0.72$  aF, that between the Au core and the source  $C_2=0.61$  aF, the fractional residual charge  $Q_0=0.2e$  and the capacitance between the gate electrode and the Au core  $C_g=99$  zF. When the Au nanoparticles with the diameter of  $5.2\pm0.5$  nm are used as the Coulomb island in a planer Au nanogap electrodes without the top-gate electrodes, an average  $C_{\rm g}$  is evaluated as 6.7 zF[5]. Comparing with this value, the obtained  $C_g$ =99 zF is 15 times larger than that comes from the planer structure SETs.

### 4. Summary

We fabricated a metal bridged top-gate electrode on Au



Fig. 2 Stability diagram of single-electron transistor with top-gate electrode at 9 K. The conductance peak lines calculated from  $C_1$ =0.72 aF,  $C_2$ =0.61 aF,  $C_g$ =99 zF and  $Q_0$ =0.2e are also shown.

nanogap electrodes by using two consecutive electron beam lithographies. With these Au nanogap electrodes and chemically synthesized Au nanoparticles, we demonstrated an operation of single-electron transistors. An introduction of top-gate electrodes increases the gate capacitance  $C_{\rm g}$ .

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