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著者(和文)	ChoiYoonYoung
Author(English)	Yoonyoung Choi
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Outline of Thesis

As an artificial intelligence (AI) era is dawning, the processing of huge amounts of information data is requested for electronic devices. In order to meet these demanding, transistors are required to have the higher performance and higher efficiency ; low power consumption, high switching speed, small size. Therefore, transistors have been improved in their performance and decreased in their size to few tens of nanometer. However, there has still been the limitation of transistor performance and scaling to sub-10nm scale. In these aspect, single-electron transistors (SETs) and molecular transistors have been suggested and developed. However, in order to realize these SETs and molecular transistors operated at room temperature (RT), the robust and ultra-fine nanogap electrodes with sub-3nm in gap separation have been strongly requested.

Accordingly, in this thesis, I have developed and established the fabrication processes of robust and ultra-fine Pt-based nanogap electrodes with sub-3nm in gap separation and sub-20nm in linewidth for the realization of SETs or single-molecular transistors by electron beam lithography (EBL) and electroless gold plating (ELGP) as mass-production possible method.

In order to fabricate the robust and ultra-fine nanogap electrodes, I have changed the base electrodes material from Au to Pt for the narrow source/drain (S/D) linewidth on 10nm scale and for the robust thermal stability against Rayleigh instability. By the optimization of nanogap design, dose exposure, and several EBL processes, ultra-fine Pt-based nanogap electrodes with 10 nm in S/D linewidth were fabricated and robust thermal stability was achieved in Chapter 2.

For obtaining sub-3nm-scale gap separation in the ultra-fine Pt-based nanogap electrodes, which is suitable for the size of single nanoparticle (NP) or single molecule for RT operable SET and single-molecular transistor, ELGP processes of ultra-fine Pt-based nanogap electrodes were developed. By the introduction of pre-treatment process and the optimization of several plating conditions, the controllability of sub-3nm in gap separation and sub-20nm in S/D linewidth over Pt-based nanogap electrodes was achieved by uniform-ELGP (U-ELGP) and hemispheric-ELGP (H-ELGP) in Chapter 3. Moreover, these ultra-fine ELGP Pt-based nanogap electrodes with sub-3nm in gap separation and sub-20nm in S/D linewidth have shown the strong durability against Rayleigh instability.

Finally, in order to check the effect of the ultra-fine Pt-based nanogap electrodes on SET operation, the chemically assembled SETs by chemisorbing Au nanoparticle (NP) and the colloidal InP@ZnS quantum dot (QD) as Coulomb island were fabricated and evaluated in their electrical properties as a function of operation temperature in Chapter 4. Consequently, the stable SET operation was achieved by large gate capacitance originated from sub-20nm-scale ultra-fine

S/D linewidth. Moreover, InP@ZnS QD SET have shown the stable switching characteristics and unique hysteresis behavior in $I - V$ characteristics.
For more detail, I summarized this thesis by every chapters as follows:

• Chapter 1: Introduction

In this chapter, I explained the requirement and necessity of nanogap electrodes for the realization of nanoscale transistor such as SETs and single molecular transistors and then, introduced several fabrication methods for nanogap electrodes. Lastly, I introduced the basic operation principle and reports of SET and molecular device and suggested objective of this thesis

• Chapter 2: Development of Fabrication Processes for Ultra-Fine Patterned Pt-based Nanogap Electrodes

For the fabrication of the robust and ultra-fine nanogap electrodes with very small gap separation and ultra-narrow S/D linewidth for large gate capacitance, Pt as base electrodes material and EBL were introduced because Pt has the higher melting point and 10^7~10^8 times smaller surface diffusion coefficient value than that of Au, and EBL is mass-production available process. By optimizing nanogap design, dose exposure condition and several EBL processes, the robust Pt-based nanogap electrodes with 10 nm in ultra-fine linewidth and with ~6nm in gap separation have been fabricated on SiO_2/Si substrates. In order to check the robustness of these ultra-fine Pt-based nanogap electrodes, several stability tests (thermal, chemical, and mechanical tests) were carried out. As results, ultra-fine Pt-based nanogap electrodes maintained their electrodes structure unchanged up to temperatures of 773 K. In contrast, ultra-fine Au-based nanogap structure could not be fabricated by using EBL process; they deformed into droplets owing to Rayleigh instability. This robust thermal stability of ultra-fine Pt-based nanogap electrodes was attributed to the low surface diffusion coefficient of Pt. Moreover, ultra-fine Pt-based nanogap electrodes have also shown the strong resistance against the chemical etching and mechanical stress.

Finally, the structural robustness of the developed ultra-fine Pt-based nanogap electrodes has been demonstrated in thermal, chemical and mechanical aspect.

• Chapter 3: Development of Electroless Au Plating (ELGP) Processes over Pt-based Nanogap Electrodes

To obtain sub-3nm in gap separation of ultra-fine Pt-based nanogap electrodes, ELGP processes were developed. For the cleaning of Pt surface before ELGP process, several pre-treatments were investigated. As results, U-ELGP and H-ELGP over ultra-fine Pt-based nanogap electrodes could be selected and controllable by pre-treatment condition. By optimization of plating solution concentration and time, sub-3nm in gap separation and sub-20nm in S/D linewidth were fabricated by self-termination mechanism. Moreover, from the cross-sectional structure analysis of ultra-fine H-ELGP Pt-based nanogap electrodes by SEM, STEM and TEM, ultra-small gap separation as less than 1nm was verified and Au growth over the Pt polycrystalline was confirmed, which cause the structural robustness of hemispheric Au structure over Pt surface. In addition, these unique hemispheric nanogap ends structure at the gaps could improve the SET operation characteristics owing to the larger gate capacitance originated from additional reduction of S/D linewidth at the gaps.

Finally, thermal stability of ultra-fine ELGP Pt-based nanogap electrodes was evaluated by comparing with the previous ELGP Au-based nanogap electrodes. The superiority of ultra-fine U-ELGP Pt-based nanogap electrodes and H-ELGP Pt-based nanogap electrodes was demonstrated obviously.

• Chapter 4: Application of Ultra-Fine Pt-based Nanogap Electrodes for Single-Electron Transistors (SETs)

In order to confirm the effect on SET operation and to explore SET with new functionality by using the developed ultra-fine Pt-based nanogap electrodes, the chemically assembled SETs by chemisorbing Au NPs and InP@ZnS QDs were fabricated and evaluated in their electrical properties as a function of operation temperature.

Firstly, I fabricated the chemically assembled SET by chemisorbing the 6 nm-sized Au NPs as Coulomb island between ultra-fine Pt-based nanogap.

As measurement results of the I_d - V_d, I_d - V_g and stability diagram characteristics of Au NP SETs as a function of operation temperature, the clear Coulomb blockades, Coulomb oscillations and Coulomb diamonds were observed, which indicating that this SET shows the stable switching operation with gate voltage. And then, theoretical I_d - V_d was fitted with the experimental I_d - V_d curve. As fitting results, theoretical I_d - V_d curve agreed well with the experimental I_d - V_d result, which means that this SET shows ideal and stable operation. From the theoretical fitting results of I_d - V_d and Coulomb diamonds, the tunneling capacitance (C_1 = 1.075 aF, C_2 = 1.074 aF, C_{g2} = 48 zF), tunneling resistance (R_1 = 0.175 MΩ, R_2 = 1.35 MΩ,) and charging energy (E_c = 36.4 meV) of Au NP SET were also evaluated.

By comparing this gate capacitance with that of previous SET result by Au-based nanogap electrodes with 40~90 nm in linewidth, the chemically assembled Au NP SET by ultra-fine Pt-based nanogap electrodes with 10 nm-scale S/D linewidth had at least 1.5 times larger gate capacitance value. From the stability diagrams result with operation temperature, the stable switching operation was demonstrated up to 200 K with large gate capacitance of 48 zF owing to 10 nm in S/D linewidth.

Secondly, in order to explore SET with new functionality by using the developed ultra-fine ELGP Pt-based nanogap electrodes, chemically assembled InP@ZnS QD SETs were fabricated by introducing the colloidal InP@ZnS QDs as Coulomb island into the robust ultra-fine ELGP Pt-based nanogap electrodes.

The fabricated InP@ZnS QD SETs exhibited the stable switching characteristic and hysteresis behavior in $I-V$ curve at 9K, which is originated from charge-trapping and detrapping effect into the defect levels in InP@ZnS QD. For the better understanding of transistor operation characteristics for InP@ZnS SET, the two-dimensional I_d and dI_d/dV_d plot as function of V_d and V_g were measured at 9K and 80K. The clear and rhombic Coulomb diamonds were obtained in the stability diagram at 9K and the stable operation of InP@ZnS SET up to 80K was demonstrated.

• Chapter 5: Conclusions

Finally, I summarized this thesis about study on fabrication processes of ultra-fine Pt-based nanogap electrodes. And then, I suggested future work to improve fabrication yield and mentioned future perspective of ultra-fine Pt-based nanogap electrodes