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Dithiol-Linked Gold Colloidal Particles Used for Fabricating Single Electron Transistors

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1. Introduction

The application of cluster chemistry to single electron device fabrication was recently demonstrated by Klein et al. [1] and Sato et al. [2]. By using an alkanedithiol, they were successful in binding cluster particles in a lithographically-formed metal gap and demonstrated Coulomb staircases in electron transport measurements through the dots of metal/semiconductor clusters. In this device configuration, the alkane chain molecules act as electron tunneling barriers while the cluster particle acts as an electron localization site. In this paper, periodic conductance oscillations due to gate biasing will be presented extending the previous electrical measurement on such a cluster-based single electron devices.

2. Linking gold particles by dithiol molecules

The device structure is shown in Fig. 1. A chain of gold colloidal particles (GCPs) was formed on a thermally-grown SiO_2 (200 nm thick) surface on a Si substrate on which the source and drain electrodes,

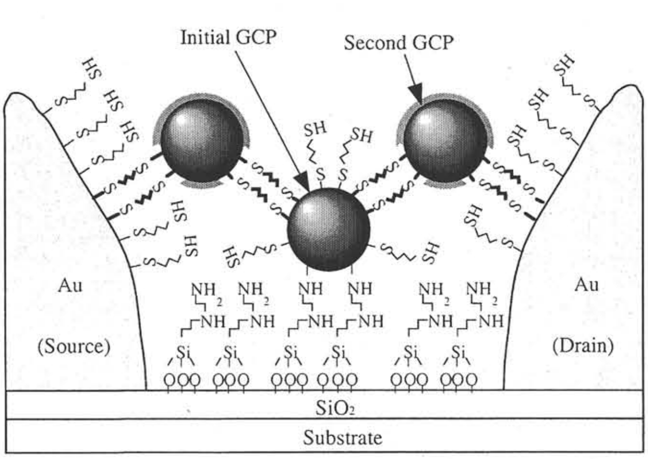
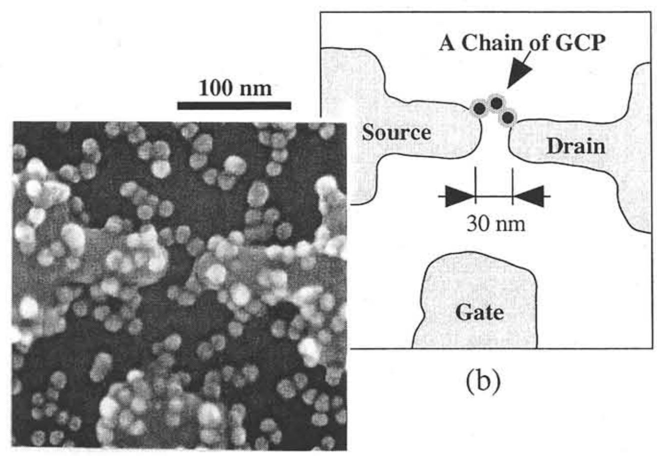


Fig.1 Device structure for cluster-based mutiple tunnel junction. The initial particle is attached to the substrate by aminosilane molecules.



(a) (b)

Fig. 2 (a) Scanning electron microscope observation of a three-dot gold colloidal particle chain incorporated in a system of source, drain, and gate metal electrodes. (b) Schematic of electrode pattern defined by electron beam lithography and the three particle chain.

with a 30-nm gap between them, were formed using electron-beam lithography.

In order to obtain the device characteristics, at least, one of the GCPs was initially deposited in the gap between the source and drain electrodes using the aminosilane method described in ref.[2]. The diameter of the particle used here was 10 nm.

Afterwards, dithiol treatment was carried out on the gold colloidal particle to induce the chaining of the particles. More precisely, the sample on which the gold particle was deposited was immersed in a 5 mM ethanolic solution of 1,6-hexanedithiol ($SH\text{-}(CH_2)_6\text{-}SH$) for ~24 hrs, and rinsed with two ethanol baths, and dried with nitrogen gun. Because of the strong affinity of sulfur for gold, the treatment replaced surface adsorbates of the gold colloidal particles with the

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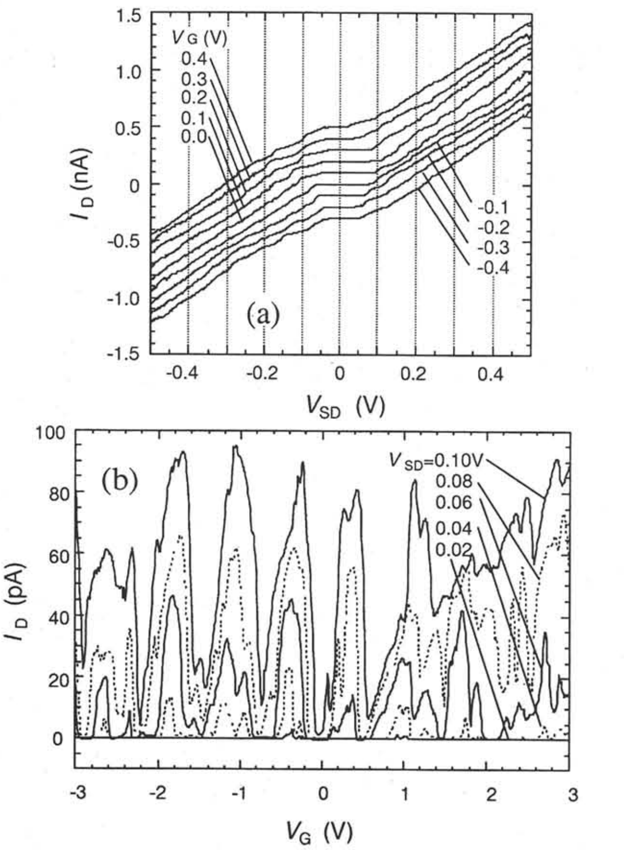

Fig. 3 (a) Drain current (I_D) vs. source-drain voltage (V_{SD}) characteristics of a three-dot GCP transistor measured at 4.2 K with various gate voltages (V_G). The I_D-V_{SD} curves are shifted by 0.1 nA. (b) Drain current (I_D) vs. gate voltage (V_{SD}) characteristics (conductance oscillations) of a three-dot GCP transistor measured at 4.2 K with various source-drain voltages (V_{SD}).

dithiol molecules. Moreover, such dithiol coating can take place also on the electrode surfaces.

Immediately after the dithiol treatment, the second GCP immersion was carried out. Because of the thiol group termination, incoming GCPs in the second immersion were immobilized on both the firstly-laid GCPs and on the Au electrodes. As a consequence, a chain of GCPs bound to the gold electrodes was formed in between the gap as shown in Fig.1.

Figure 2 (a) shows a scanning electron micrograph of a device of which source-drain gap is bridged by a chain of three GCPs, while Fig. 2(b) shows an electrode patern defined by lithography.

3. Electrical characteristics measurement

The I_D-V_{SD} characteristics were measured with different fixed gate voltages ranging from V_G = -0.4 to 0.4 V at 4.2 K. The observed I_D-V_{SD} characteristics exhibited Coulomb gaps and Coulomb staircases as shown in Fig.3 (a). Squeezing of the Coulomb gap was observed with increasing magnitude of the gate voltage V_G. The Coulomb gap at its maximum reached to ~150 mV.

The measurement of I_D as a function of V_G was also carried out at fixed V_{SD} values ranging from 0.02 to 0.1 V. As shown in FIG. 3 (b), the observed I_D exhibited periodic changes within the applied gate voltage range, which is characteristic of single electron tunneling across a multiple tunnel junction.

A 10-nm metal sphere embedded in a uniform dielectric medium with a dielectric constant value similar to that of an alkane chain monolayer (i.e., ε_r = ~ 2.8) [3] gives a self-capacitance value of ~ 3 aF. The micrographical examination of the device clearly showed that three 10-nm particles formed a quasi one-dimensional current path across the source-drain gap (see Fig.2); each particle is sandwitched by two tunnel junctions connecting it either to adjacent particles or one of the electrodes. Therefore a good approximation for junction capacitance C_J attached to each Au particle sould be a half the self-capacitance value given above. Using such junction capacitance value C_J, one can calculate the Coulomb gap value for the three-dot (four-junction) system assuming that all the junction capacitances C_J are equal[4]. The low-impedance Coulomb gap ΔE for the four-junction system is calculated to be $\Delta E \sim (N - 1)(e / 2\ C_J)$ = 160 mV, where N is the number of junctions (i.e., N = 4) and C_J = 1.5 aF (= 3aF / 2). This value is in reasonable agreement with the value $\Delta E \sim$ 150 mV obtained from the measurement shown in FIG. 3 (a). The agreement between the experiment and the simulation suggests that the observed current was a manifestation of sequential single electron tunneling through the three-dot chain of 10-nm gold particles.

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