

Large Negative Resistance Property Observed in 3-D Network of DNA and Gold Nanoparticle Formed by DNA Mediated Self-organization

Y. Fujii, T. Shimizu, M. Hosoda, G. Wu, S. Huang, H. Sakaue, T. Takahagi, and S. Shingubara

Graduate School of ADSM, Hiroshima University Kagamiyama 1-3-1, Higashi-hiroshima, 739-8526, Japan

Phone: +81-824-22-7111 Fax: +81-824-24-0709 E-mail: fujii@hiroshima-u.ac.jp

1.Introduction

DNA-mediated assembly of nanometer scale structures may have a profound effect in fabricating nanoelectronic devices and biosensors [1]. We recently developed a method to form three-dimensional (3-D) network of gold nano particles by DNA hybridization using complementary thiol-capped DNA oligonucleotides [2]. In this study, we formed a 3-D network of gold nanoparticle and DNA in between a narrow gap of gold split-electrodes, and investigated electrical conduction characteristics of this structure. We observed an enhanced negative resistance property for some samples, although there were large sample-to-sample fluctuations.

2.Experimentls

We formed gold split-electrodes with a gap of 40 nm by electron beam lithography and lift-off technique. We used two complementally single chained DN; one end of which was chemically bonded to thiol (S-H). These were thiol-5'-GGTCGGCACA-3'(GG-10), and thiol-5'-TGTGCCGACC-3'(CC-10). Lengths of GG-10 and CC-10 were almost 10nm. At hybridization conditions (typically pH=7.0, 40 °C, 48 hours), CC-10 and GG-10 combined each other to form double helix structure, which is the key issue of DNA mediated self-organization. Gold nanoparticles with average diameter of 8 nm were prepared by wet chemical reduction of chloroauric acid (HAuCl₄) with reducing agents of trisodium citrate and tannic acid.

Fabrication procedure of 3-D network of gold nanoparticle and DNA in between gold split gate is schematically shown in Fig.1. At first, surfaces of gold electrode were covered by CC-10 in a self-organizing manner due to strong interaction between Au and S atoms. On the other hand, two types of gold nanoparticle colloidal solutions were prepared: gold nanoparticle covered with GG-10, and that covered with CC-10. Then each gold colloidal solution was poured on the substrate with gold split-electrodes sequentially, and DNA hybridization was carried out.

We fabricated more than 30 samples, and there were a few samples that exhibited clear negative resistance characteristics at liquid helium temperature (4.3 K). A typical example is shown in Fig.2. Current was suppressed when an absolute value of voltage was smaller than 1.1 V.

Then exponential increase of the current against voltage was observed. It should be notified that there was a huge negative resistance peak at 3.2 V. In the differential current-voltage relationship (dI/dV), we identified another broad negative peak around 4.2V. In the range of negative voltage, there was also a broad negative peak around -4.2V, and furthermore a very small negative peak existed at -2.6 V. We may say that a resonant tunneling phenomena with asymmetric barriers occurred in this structure. A SEM plan view micrograph (Fig.3) showed that there were gold nanodots encapsulated by DNAs on the surface of gold split-electrodes. There was a cluster of DNA-nanoparticle-complex with size of 20 x 50 nm² at the gap between gold electrodes. A model of resonant tunneling is shown schematically in Fig.4. There is a quantum dot with two energy levels whose energy splitting is about 1.0 eV, sandwiched with asymmetric barriers. The most plausible origin of the quantum dot is a very small gold cluster whose diameter is smaller than 3nm. Li et. al reported an evidence of semiconductor gold cluster with a huge energy levels splitting [3], and Zheng et al. observed blue photoemission from gold clusters [4]. Inclusion of such very small gold clusters is possible by wet chemical synthesis, and DNA is likely to act as a tunnel barrier in the present system.

3.Conclusions

A large negative resistance was observed in the nanometer scale 3-D Network of gold nanoparticle and DNA. The origin of negative resistance is considered to be a resonant tunneling phenomenon occurred at the ultra fine gold cluster sandwiched by asymmetrical barriers. These results strongly suggest a usefulness of DNA-mediated self-assembly of nanomaterials for active electronic device fabrications.

References

- [1] R.Bashir, Superlattice and Microstructures, **29** (2001) 1.
- [2] T.Takahagi, G.Tsutsui, S.Huang, H.Sakaue, and S.Shingubara, Jpn.J.Appl.Phys. **40** (2001) L521.
- [3] J.Li, X.Li, H.-J.Zhai, L.-S. Wang, Science **299** (2003) 864.
- [4] J.Zheng, J.T.Petty, and R.M.Dickson, J.Am.Chem.Soc. **125** (2003) 7780.

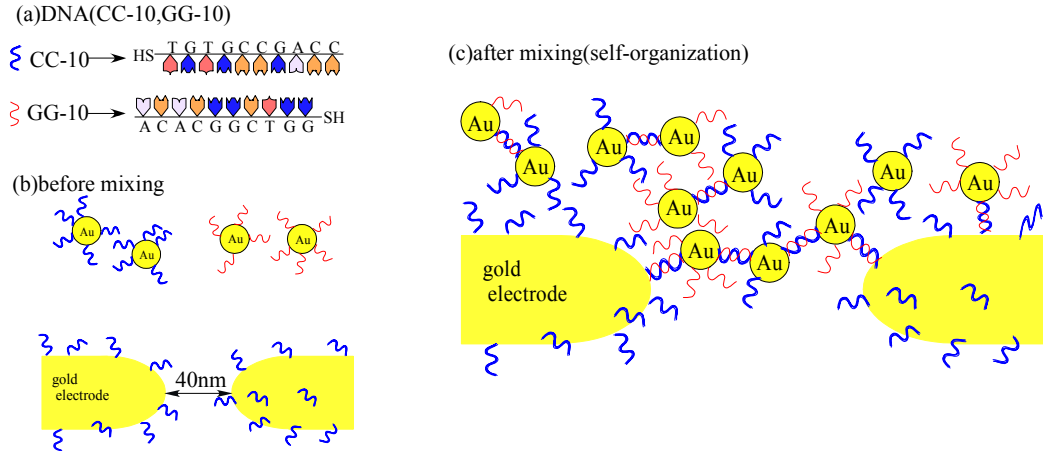


Fig.1. (a) Two types of DNA: CC-10, GG-10 (b) Preparation of two types of gold nanoparticle colloidal solutions (covered with CC-10 or GG-10), and gold electrodes covered with CC-10. (c) After DNA hybridization, 3-D network of Au nanoparticle and DNA was formed between gap of electrodes.

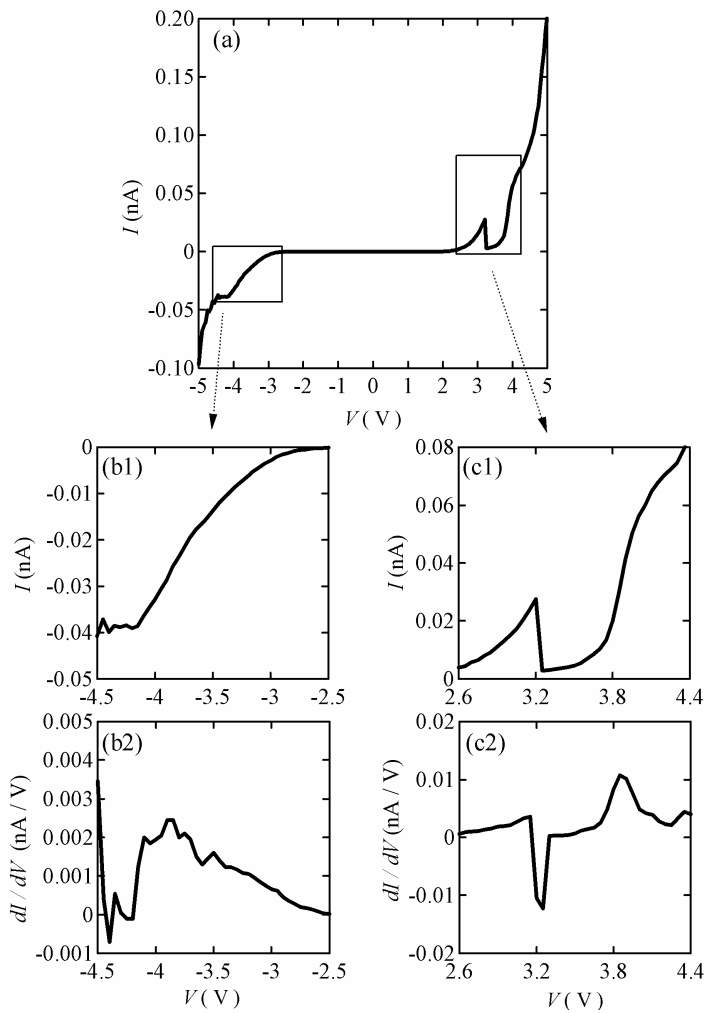


Fig.2. (a) Current-Voltage characteristics of the sample (at 4.3K). Asymmetric large negative resistance was observed. (b2) ,(c2); Differential current-voltage relationship (dI/dV). There was a broad negative peak around 4.2V in (c2). A huge negative resistance peak at 3.2V was observed in (c1).

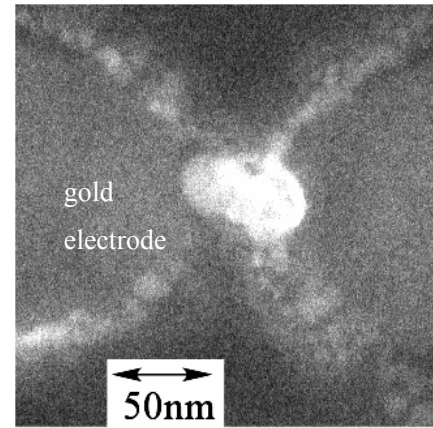


Fig.3. SEM plan-view image of the sample after DNA hybridization. There was a cluster of DNA-nanoparticle complex with size of $20 \times 50 \text{ nm}^2$ at the gap between gold electrodes.

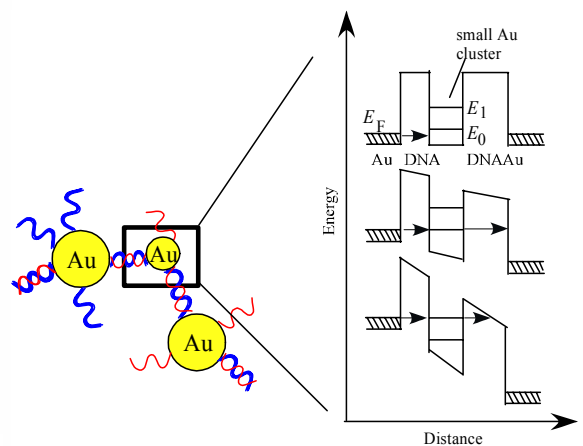


Fig.4. A model of resonant tunneling. There is a very small gold cluster, sandwiched with asymmetric barriers. Energy splitting between two level is about 1.0 eV.