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BRIEF PAPER

Dielectrophoretic Assembly of Gold Nanoparticle Arrays Evaluated in Terms of Room-Temperature Resistance

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SUMMARY Gold nanoparticles (GNPs) are often used as island electrodes of single-electron (SE) devices. One of technical challenges in fabrication of SE devices with GNPs is the placement of GNPs in a nanogap between two lead electrodes. Utilization of dielectrophoresis (DEP) phenomena is one of possible solutions for this challenge, whereas the fabrication process with DEP includes stochastic aspects. In this brief paper, we present our experimental results on electric resistance of GNP arrays assembled by DEP. More than 300 pairs of electrodes were investigated under various DEP conditions by trial and error approach. We evaluated the relationship between the DEP conditions and the electric resistance of assembled GNP arrays, which would indicate possible DEP conditions for fabrication of SE devices.

key words: *single-electron effect, Coulomb blockade, histogram, tunnel junction*

1. Introduction

Single-electron (SE) devices [1], which are one of the candidates for the "beyond CMOS" technologies [2], are composed of several tiny tunnel junctions and tiny island electrodes. Since the report by Schönenberger *et al.* [3], gold nanoparticles (GNPs) have been often used for island electrodes in SE devices because of their small sizes and chemical stability. In regard to lead electrodes, besides a tip of a scanning probe microscope, two metal electrodes placed across a nanogap are commonly used for the source and drain electrodes [4]–[8]. For such a pair of two electrodes, it is a technical challenge to embed one or a few GNPs in the nanogap.

Utilization of dielectrophoresis (DEP) phenomena is one of the techniques to convey GNPs into a nanogap [9]–[16]. A non-uniform electric field exerts a dielectrophoretic force on a dielectric particle in the field. The time-averaged DEP force $\langle \boldsymbol{F}_{\mathrm{DEP}}(t) \rangle$ is expressed as

$$\langle \boldsymbol{F}_{\mathrm{DEP}}(t) \rangle = 2\pi\varepsilon_{\mathrm{m}} a^3 \mathrm{Re}\left[K(\omega)\right] \nabla |\boldsymbol{E}_{\mathrm{rms}}|^2 \quad (1)$$

where ε_{m}, a, $K(\omega)$, and $\boldsymbol{E}_{\mathrm{rms}}$ are the permittivity of the medium, the radius of the nanoparticle, the Clausius-Mossotti factor, and the rms value of the electric field, respectively [17].

So far, several groups have reported SE devices in which GNPs are assembled in an electrode gap by using the DEP force [18]–[20]. Parameters related to DEP, such as the voltages, frequencies, layouts of electrodes, etc., have been also investigated [17], [22], [23].

We have also tried to fabricate SE devices using DEP [21]. Some devices exhibited clear characteristics of the Coulomb blockade and Coulomb oscillation. On the other hand, we have found that a wide variety of GNP arrays are realized under the nearly same DEP conditions. It is no wonder that we do not obtain the identical arrays made of a small number of GNPs since the microscopic DEP conditions are not static. For example, the positions of all GNPs in a colloidal solution are changing at every moment. GNPs attached on the electrodes simultaneously change the electric field around the gap. That is, from the viewpoint of SE devices, the fabrication process utilizing the DEP force includes stochastic aspects.

In this brief paper, we present our experimental results on electric resistance of GNP arrays assembled by DEP. Although the electric resistance of GNP array was measured at room temperature, it was good predictor of electric properties at low temperature. More than 300 pairs of electrodes were investigated under various DEP conditions by trial and error approach. Although the experimental conditions were not strictly systematized, we evaluated the relationship between the DEP conditions and the electric resistance of GNP arrays, which would indicate possible DEP conditions for fabrication of SE devices.

2. Experiments

Samples were prepared as follows. First, NiCr/Au electrodes were made on a Si/SiO_2 substrate by using e-beam lithography and a shadow evaporation technique. The thicknesses of the first and second evaporation of NiCr/Au layers were 5/20 and 5/45 nm, respectively. The layout of electrodes is illustrated in Fig. 1. Typically, 7 pairs of electrodes were fabricated on a sample. A part of samples were immersed in a 5 mM ethanol solution of either 1,10-decanedithiol or 1,4-butanedithiol for 18 hours at room temperature, which resulted in SAM coating on the electrodes.

Next, the DEP process with a colloidal citric solution of GNPs was conducted. The diameter and density of GNPs

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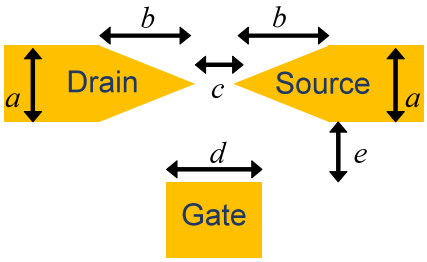

Fig. 1 Schematic illustration of electrodes (top view). An AC voltage was applied between the source and drain electrode. The dimensions are as follows. *a*: 800 nm, *b*: 1.2 μm, *c*: 100–200 nm as the target value (actually fabricated in the range of 100–400 nm), *d*: 500–900 nm, *e*: 0–1 μm.

were 30 nm and 1.8×10^{11} /mL, respectively. For some experiments, the surface of GNPs were modified by mixing the GNP colloidal solution and a 0.1 mM ethanol solution of 1-decanethiol with the volume ratio of 400 μL and 200 μL. After dropping 10 μL of gold colloidal solution on a sample, an AC sinusoidal voltage of 1–10 V_{p-p} (V_{DEP}) and 100 Hz–10 MHz (f_{DEP}) was applied across two electrodes (source and drain) for 2 s–10 min (T_{DEP}). The voltage waveform was monitored on an oscilloscope. It should be noted that we conducted the DEP process one by one for all electrode pairs on the sample with one drop of GNP colloidal solution. Because of solvent evaporation, the GNP density was not kept constant during the DEP processes even on the same sample. After the DEP process, the sample was rinsed in pure water and then dried with N_2 gas blow.

GNP arrays were evaluated by three methods, SEM observation, electric resistance measurement at room temperature, and measurement of *I*–*V* characteristics at low temperature.

We observed all electrode pairs by using an FE-SEM. From SEM images in a 500×500 nm^2 region near the electrode gap, we extracted the area occupied by GNPs. This GNP area (S_{GNP}) is an evaluation index of GNPs assembled by the DEP process.

Then, at room temperature, the resistance values of all electrode pairs were measured by a resistance meter with self-build terminals. The effective measuring range was between 10 kΩ and 28 GΩ. It was empirically observed in our preceding experiments that the electrode pairs having the resistance values out of this range seldom exhibited the Coulomb blockade. Thus, the room-temperature resistance (R_{RT}) is another evaluation index.

Finally, *I*–*V* characteristics were measured in a liquid helium bath. Due to the experimental costs, 25 electrode pairs were evaluated.

3. Results and Discussion

Figure 2 shows an example of the SE devices fabricated using the DEP process, where surface-modified electrodes were used with the DEP conditions of f_{DEP} = 1000 kHz, V_{DEP} = 4 V, and T_{DEP} = 10 s. It is found in Fig. 2 (a) that GNPs are assembled in the electrode gap. S_{GNP} was

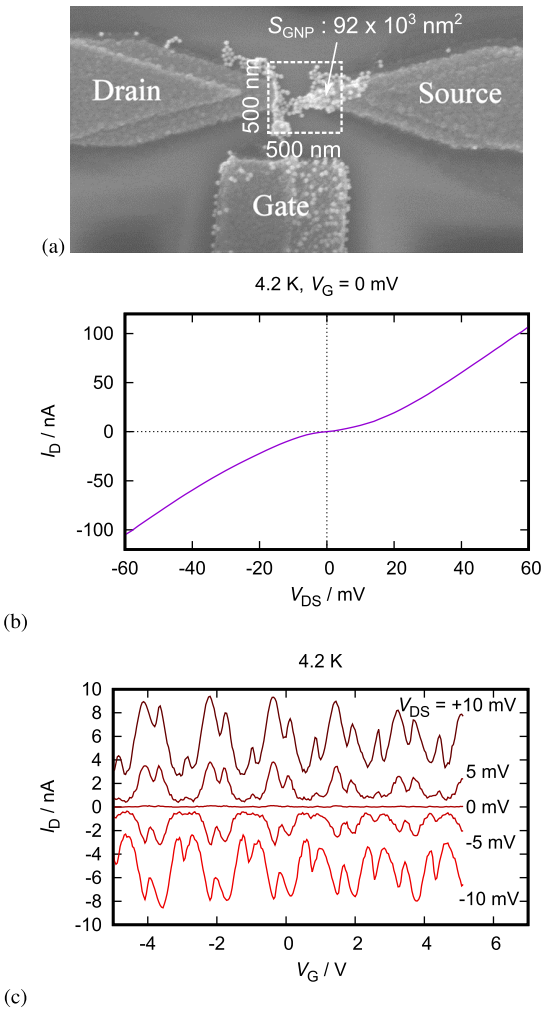

Fig. 2 Example of the fabricated devices (f_{DEP} = 1000 kHz, V_{DEP} = 4 V, T_{DEP} = 10 s). (a) SEM image around the electrode gap. S_{GNP} and R_{RT} are 92×10^3 nm^2 and 360 kΩ, respectively. (b) Drain current (I_D) vs. drain–source voltage (V_{DS}) characteristics at 4.2 K (c) I_D vs. gate voltage (V_G) characteristics at 4.2 K.

determined to be 92×10^3 nm^2, whereas R_{RT} was 360 kΩ. Electric properties at liquid helium temperature shown in Figs. 2 (b) and 2 (c) demonstrate clear Coulomb blockade and Coulomb oscillation.

As described in the introduction, the DEP process is rather stochastic. In other words, while we succeeded to fabricate SE devices as shown in Fig. 2, we also had electrode pairs that exhibited electric characteristics like a linear resistor or an insulator. To find out better DEP conditions for fabrication of SE devices, we employ not rigorous but somewhat statistical approach. R_{RT} is used for the main index. It is noted again that electrode pairs having R_{RT} between 10 kΩ and 28 GΩ, which is hereafter referred to as the intermediate R_{RT}, are empirically expected to work as SE devices. The numbers of electrode pairs classified by R_{RT} and the electric characteristics measured at 4.2 K are tabulated in Table 1, which supports our empirical assumption.

The first analysis is the f_{DEP} dependence of R_{RT} shown

Table 1 Number of electrode pairs classified by R_{RT} and the electric characteristics measured at 4.2 K.

R_{RT}	Electric properties at 4.2 K		
	linear	non-linear	open
< 10 kΩ	1	0	0
10 kΩ – 28 GΩ	0	13	0
> 28 GΩ	0	2	9

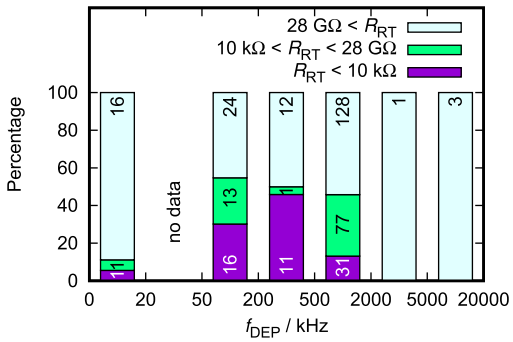

Fig. 3 R_{RT} distribution classified by f_{DEP}. Numerals in bins represent the number of corresponding electrode pairs. Note that the f_{DEP} regions on the horizontal axis are not equally divided.

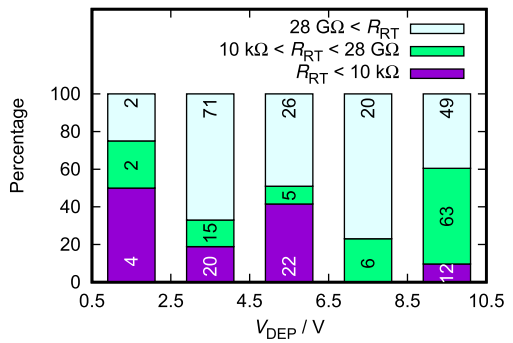

Fig. 4 R_{RT} distribution classified by V_{DEP} for f_{DEP} > 20 kHz. Numerals in bins represent the number of corresponding electrode pairs.

in Fig. 3, where all results for the fabrication conditions described in the previous section are included. It is found that the intermediate R_{RT} is obtained for 50 < f_{DEP} < 2000 kHz. For f_{DEP} < 20 kHz, 16 among 18 samples have R_{RT} larger than 28 GΩ, which allow us to exclude the results for f_{DEP} < 20 kHz hereafter. On the other hand, although we obtained no samples having the intermediate R_{RT} for f_{DEP} > 2000 kHz, we include them for the further analyses because the number of samples are too small to exclude them.

Next, dependence analyses of R_{RT} on V_{DEP}, T_{DEP} and the product of $T_{DEP}V_{DEP}$ are respectively presented in Figs. 4, 5, and 6. It is seen that the intermediate R_{RT} is efficiently obtained under the DEP conditions of V_{DEP} > 8.5 V, T_{DEP} > 50 s, and $T_{DEP}V_{DEP}$ > 200 Vs. The upper limits for these parameters cannot be determined due to the limited number of samples.

Finally, relationship between R_{RT} and S_{GNP} is presented in Fig. 7. It is found that larger S_{GNP} is effective to make R_{RT}

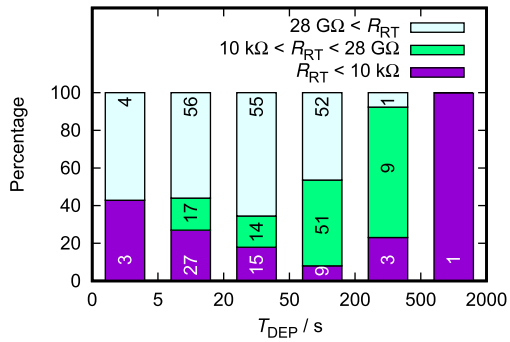

Fig. 5 R_{RT} distribution classified by T_{DEP} for f_{DEP} > 20 kHz. Numerals in bins represent the number of corresponding electrode pairs. Note that the T_{DEP} regions on the horizontal axis are not equally divided.

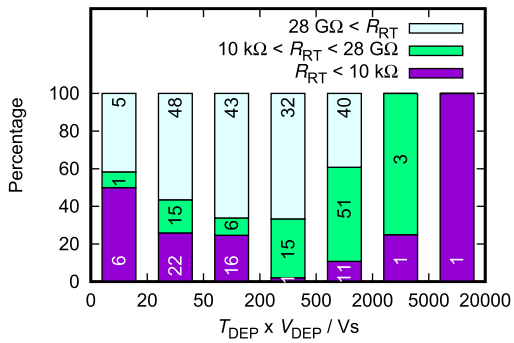

Fig. 6 R_{RT} distribution classified by the product of T_{DEP} and V_{DEP} for f_{DEP} > 20 kHz. Numerals in bins represent the number of corresponding electrode pairs. Note that the $T_{DEP}V_{DEP}$ regions on the horizontal axis are not equally divided.

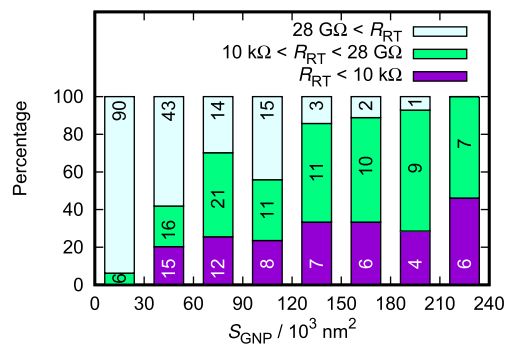

Fig. 7 R_{RT} distribution classified by S_{GNP} for f_{DEP} > 20 kHz. Numerals in bins represent the number of corresponding electrode pairs.

smaller than 28 GΩ. A threshold exists near 30×10^3 nm^2, above which electrode pairs have finite resistance. Another threshold seems to be near 120×10^3 nm^2, above which insulative states almost disappear. Such S_{GNP} dependence of R_{RT} agrees with the natural relationship between the electrical connection and the amount of assembled GNPs.

4. Conclusion

We presented our experimental results on electric resistance of GNP arrays assembled by DEP. More than 300 pairs

of electrodes were investigated under various DEP conditions by trial and error approach. We evaluated the relationship between the DEP conditions and the electric resistance of GNP arrays, which would suggest possible DEP conditions for fabrication of SE devices. That is, the conditions of $f_{\mathrm{DEP}} > 20\,\mathrm{kHz}$, $V_{\mathrm{DEP}} > 8.5\,\mathrm{V}$, $T_{\mathrm{DEP}} > 50\,\mathrm{s}$, and $T_{\mathrm{DEP}} V_{\mathrm{DEP}} > 200\,\mathrm{Vs}$ provide rough guidelines.

The results presented above were obtained in our experiments by trial and error. For further statistical evaluation of the DEP process, experiments should be organized well. However, although they included trivial conclusions, these results are the first step for combining the DEP conditions and electric characteristics of GNP arrays.

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