Nanogap size control by room-temperature annealing

T. Hayashi and K. Muraki
INTT Basic Research Laboratories, NTT Corporation, 3-1 Morinosato-Wakamiya, Atsugi, Kanagawa 243-0198, Japan
E-mail: toshiaki@nttbrl.jp, TEL: 046-240-3447, FAX: 046-240-4723

1. Introduction
Field-effect semiconductor lateral quantum dots (QDs) have provided a suitable stage to study a wide variety of interesting quantum transport phenomena such as the Coulomb blockade and the Kondo effects that occur in the weak- and strong-coupling regimes, respectively. The main advantage of such devices is that the tunnel couplings of the QD to the source/drain leads can be readily controlled by tuning the corresponding gate voltages. On the other hand, single-molecule QDs, which form a new class of QD systems, consist of a single molecule sitting in a nanogap between two metallic electrodes and a gate electrode nearby to tune the electrostatic potential of the molecule. In many studies, the nanogaps are created by using controlled electromigration followed by self-breaking of a thin Au nanowire. As a result, there is not much controllability over the final nanometer-scale structure of the device nor the tunneling rates dominated by the spatial distance between the molecule and the electrodes.

In this work, we introduce a new scheme to improve the flexibility in the molecular device fabrication. The scheme is based on the fact that self-breaking is driven by the surface tension built up near the atomic chain and therefore continues even after the rupture of the chain. The obtained Au nanogap continuously widens at room temperature. We demonstrate that repeating $I$-$V$ measurement at low temperatures and thermal annealing at room temperature allows us to reduce the tunneling rates in a controlled way.

2. Experiment
Starting Au nanowires (15-nm thick, 80-nm long, and 400-nm long) was defined by electron beam lithography on a thin Al$_2$O$_3$ insulating layer covering a patterned Al backgate fabricated on a semi-insulating GaAs substrate (Fig. 1). After the sample was immersed in a C$_{60}$ solution in toluene (50 μM) for 5 min. and dried, controlled electromigration with active feedback of the source-drain voltage was performed for each of the nanowires at room temperature in vacuum to narrow it at one part (Fig. 2). Once the conductance of the nanowire has reduced to 500 μS, the feedback and the time evolution of the conductance was monitored at low source-drain voltages (±50 mV). In the device shown in Fig. 3, the reduction in the conductance stopped around 500 μS at room temperature, so that the temperature was raised to 330 K to facilitate the self-breaking of the remaining Au atomic chain. After the conductance dropped below the quantized conductance (77 μS), the temperature was lowered to 6 K in 20 min. to measure the $I$-$V$ characteristics.

Figure 4 compares the $I$-$V$ curves of the same device at 6 K taken at the first cool down [Fig. 4(a)] with those taken after several cycles between room-temperature annealing and low-temperature measurement [Fig. 4(b) and 4(c)]. In each set of data, gate-dependent $I$-$V$ curves typical of Coulomb blockade are observed, signaling the trapping of a single molecule in the nanogap. At the first cool down, the saturation current for the positive and negative biases is 28 nA and 12 nA, respectively [Fig. 4(a)]. If the tunnel barriers are assumed to be symmetric, the tunneling rates are estimated to be $\Gamma \sim 150$ GHz from the saturation current of $I = 12$ nA (i.e. $I = e\Gamma/2$). After repeating room-temperature annealing (for example, for a few hours) several times, the tunneling rates becomes progressively smaller [Fig. 4(b) and 4(c)]. Similar analysis yields tunneling rate of $\Gamma \sim 15$ and 1.5 GHz for the data shown in Fig. 4(b) and 4(c), respectively. After further annealing at 330 K for 1 hour, the current eventually disappeared completely (data not shown), implying that the nanogap became too large. We find that while the overall shape of the Coulomb diamond is unchanged after the thermal annealing, its position shifts in the gate voltage, which means that there is a change in the built-in potential of the QD.

3. Summary
In summary, we have proposed and demonstrated a new scheme of thermal annealing to control tunneling rates of single-molecule quantum dots. This technique will open up an opportunity to adjust one of the most important device parameters for single molecule devices.

References

**Fig. 1** Schematic of device structure.

**Fig. 2** $I/V$-$V$ characteristic of Au nanowire during controlled electromigration at room temperature in vacuum.

**Fig. 3** Time evolution of conductance of Au nanowire at room temperature. At 1.4 hours later, the temperature was set at 330 K to accelerate self-breaking. The conductance was monitored at low bias voltages (+/- 50 mV)

**Fig. 4** Evolution of I-V curves at 6 K. (a) first cool down just after nanogap formation, (b) after several thermal annealing at room temperature from (a), (c) after several thermal annealing at room temperature from (b).