High-temperature single-electron transistor based on a gold nanoparticle

S.A. Dagesyan1*, A. S. Stepanov2, E. S. Soldatov1, G. Zharik1
1. Lomonosov Moscow State University, faculty of physics, Moscow, Russia,
2. Scobeltsyn Institute of Nuclear physics, Moscow, Russia.

ABSTRACT

Molecular single-electron transistors based on small (2 - 4 nm) gold nanoparticles were fabricated using an electron-beam lithography and the electromigration method. Electrical characteristics of the obtained transistors were measured at 77 and 300 K. The characteristics show that the regime of a correlated tunneling of electrons was realized at these high for the process temperatures.

Keywords: single-electronics, molecular electronics, electromigration, gold nanoparticle, correlated tunneling.

1. INTRODUCTION

Continuing progress in miniaturization of basic components in electronic circuits allows searching for new physical phenomena as a basis for a future nanoelectronics. One of such effects taking place on a nanometer scale is a correlated tunneling of electrons [1]. The most simple device that allows observation of the effect is a single-electron transistor [2]. It consists of two electrodes with a central conducting island between them separated from each other by tunnel junctions and a third electrode capacitively coupled with an island. The effect can be destroyed by thermal or quantum fluctuations. So the transistor can operate only if the following condition is performed:

\[ \frac{e^2}{2C} \gg k_B T_{op} \]

where C is total capacitance of the island, \( T_{op} \) is operating temperature. It's clear that a room temperature application of the device requires an extremely low capacitance value (\( 10^{-19} \text{ - } 10^{-18} \text{ F} \)). It means that the island diameter must be less than 3 nm, i. e. it must be a molecular scale object.

A single-electron transistor operating at room temperature was demonstrated earlier [3], but it was made only using STM tip. That realization is not applicable for practical needs. So creating of a planar single-electron transistor with a high operating temperature (room temperature) is still the unsolved problem. The modern planar industrial technology possibilities in forming nanostructures are limited and they are not enough to solve this problem now. It is necessary to create electrodes (source and drain) with the gap less than 5 nm between them to obtain a single-electron transistor with a high operating temperature (\( T_{op} > 77 \text{ K} \)). At the same time nowadays there are several techniques of creating such gaps in the labs: the mechanically controllable breaking junction (MCBJ) technique [4], the electrochemical reduction of a previously formed big gap [5], the electromigration method [6] etc. They provide a formation of extremely narrow gaps (until 1 nm), but the most works in this field are devoted to researches of delicate quantum details of the electron transport through a single molecules taking place at very low temperatures (4 K and below). For example see magnetoresistive effects [7], single-photon electroluminescence of molecules [8] etc.

The goal of this work was producing of single-electron molecular transistors and researching of their properties at high temperatures (77 and 300 K). Small (2 - 4 nm) gold nanoparticles functionalized by octanethiols from Sigma Aldrich had been chosen as models of molecular objects. An electron beam lithography combined with the electromigration technique was used to form electrodes of molecular transistors.

*dagesyan@physics.msu.ru; cryolab.physics.msu.ru;
2. SAMPLE PREPARATION

The common route of experimental samples creating is as follows. Thin and narrow gold nanowires were created on the insulated substrate (Fig. 1b). The nanowires were broken using electromigration process. Several nanometer gaps were formed as a result of the process (Fig. 1c). The presence of a gap was established by the absence of the electrical conductance at low voltages. Then gold nanoparticles were deposited at the substrate from a solution (Fig. 2a). Some of them were found in the gaps (Fig. 2b). The presence of a nanoparticle in a gap was established by significant increasing of the gap conductivity.

The 3x3 mm monocrystalline silicon substrates were a base for the samples. They were insulated from future structures using 400 nm SiO₂ layer. It was deposited with magnetron sputtering in the mixture of Argon and Oxygen plasma at 1.5 Pa pressure. Then the substrate was spin-coated by PMMA A2 resist at 3500 rpm. A pattern was formed in resist mask after an electron-beam deposition and the developing in water and isopropyl alcohol mixture (in the ratio 7:93). The next step is thermal sputtering of thin gold film (14 nm) with 2 nm buffer layer of Al₂O₃. The aluminum oxide is necessary for gold adhesion to a SiO₂ substrate [9]. As a result of a lift-off procedure the following structures were formed: micrometer wide wires leading to the future transistors (Fig. 1a), 60 nm wide gold nanowires (Fig. 1b), a side-gate for each nanowire (Fig. 1b). A distance between a nanowire and its side gate is about 150 nm.

Nanowires were controllably broken using the well known electromigration process [6], i.e. by passing a high density current through them. A 3 – 5 mA current is necessary to initiate the electromigration for our nanowires geometry. The algorithm of nanowires breaking consists of two general steps. The first one is a measuring of a nanowires resistance R₀ at low current (~ 0,1 mA). The second step is a gradual increasing of a current up to several milliamps at careful control of a process by feedback system [10]. This system turns off the current as soon as the electromigration activated, i.e. when a nanowire resistance becomes significantly more than R₀. Then a new iteration starts from the first step. Our hardware provides 20 – 25 μs reaction time for feedback control. It is significantly less than the typical time (Δt ~ 1 ms) of a gold film reorganization during an electromigration [11]. It allows a very sensitive control of the process and soft, gradual evolution of the nanowire structure. About 300 – 500 iterations are usually required until the resistance of a nanowire reaches a value of about 2 kOhm. At this resistance the whole process is stopped and we have a quantum wire connecting two electrodes. Then this quantum wire created as the result of electromigration process spontaneously breaks itself during several hours because of a relaxation of stresses collected in the metal film during it preparation process including electromigration. This last stage is called self-breaking [12].

As a result a several nanometer gap forms in the gold nanowire. Scanning electron microscope used in the work (SUPRA 40) provides to measure the gap size with accuracy of about 2 nm. But for gaps less than 2 nm it doesn't provide even to determine the existence of the gap. So the current-voltage characteristics of the formed gaps were measured after the breaking procedure. The resistance values of the obtained gaps at 77 K appeared to be undistinguishable from a leakage of our measurement setup (more than 500 GOhm).
Fig. 1. SEM images of the experimental samples: (a) gold wires leading to the future transistors; (b) nanowire before the electromigration process; (c) the (1 – 3) nm gap formed after the electromigration process

We found out that an scanning electron microscope imaging of the gaps dramatically changes their electrical characteristics: the conductance of gaps increases and can vary in wide range between 100 kOhm and 100 GOhm after an electron microscope imaging. Most likely it is a result of contamination under an electron beam. That's why all SEM images presented in this work are images of test structures.

Gold nanoparticles with 2 - 4 nm diameters functionalized with octanethiols had been chosen for deposition. Nanoparticles were placed into gaps using a self-assembling from the toluene solution. A concentration of the solution was chosen empirically by diluting the initial solution for maximize probability to find only single nanoparticle in the gap (Fig 2a,2b). The obtained concentration value is $7 \times 10^{-6}$ g/ml. Nanoparticles deposition procedure was as follows. The diluted solution was agitated by ultrasound to get a homogeneous nanoparticles suspension. Then a sample was held into the solution during 1 hour. In the case of nanoparticles absence in all gaps the procedure was repeated. The presence of a nanoparticle in the gap was established by electrical measurements at liquid nitrogen temperature. Specific features of electrical characteristics allowing it are presented in the “Electrical measurements” section.
3. ELECTRICAL MEASUREMENTS

Electrical measurements were made using a sample holder adapted for low temperature measurements. The holder was placed into liquid nitrogen to achieve 77 K when it was required. During the measurements a sample was in vacuum at $p \approx 1$ mbar to avoid condensation on it. Keithley 6487 picoammeter and voltage source was used both to set source-drain voltage and to measure current. Keithley 230 programmable voltage source was used to set the gate voltage.

There are several specific features of electric characteristics allowing a suggestion about the presence of a nanoparticle in the gap. The first one is a significant change of I-V curves in comparison with the same curves before the deposition. The second feature is a presence of a Coulomb blockade (Fig. 3a), i.e. a suppression of a current at low voltages. The third feature is a non-monotonic dependence of a current on a gate voltage (Fig. 3b).

The electrical characteristics of those gaps that presumably contained nanoparticles were measured more detailed at 77 K. Such gaps were characterized by measuring a bias current (I) as a function of both drain ($V_d$) and gate voltage ($V_g$). Using this data 2D stability diagrams ($I(V_d,V_g)$) were constructed (Fig. 3c). Coulomb diamonds can be seen on diagrams clearly demonstrate a single-electron character of an electron transport. The maximal obtained value of a Coulomb blockade that corresponds to the charging energy of the nanoparticle is almost 300 mV. Such energy corresponds to the electric capacitance of the island $C \approx 5 \times 10^{-19} \text{ F} = 500 \text{ zF}$. It gives an estimation of particle diameter $d \sim 2 \text{ nm}$ in the assumption that a mutual capacitance of the island and each electrode is approximately equal to the self capacitance of the island. This estimation is in a good agreement with the size of used nanoparticles. This is another confirmation of the fact that it is the nanoparticle working as a transistors island.

Fig.2 a) Gold nanoparticles deposited on a gold surface with an optimal dilution concentration b) Typical view of a nanogap after a successful deposition
The 300 mV charging energy is more than values available in literature for the similar system [13]. This result was achieved by using the smaller nanoparticles. It allows investigation of single-electron systems potential for very practically important high-temperature applications. This value of charging energy is already much more than the energy of thermal fluctuations at 300 K. So the electrical characteristics of the transistor were measured at room temperature. The current-voltage characteristic of the transistor at 300 K is presented on the Fig. 3d. Such behaviour is typical for single-electron systems at high fluctuations level [1]. An asymptotic parts of the IV-curve extrapolation to the x-axis gives the islands charging energy value. The value obtained for this sample at a room temperature (eV ≈ 200 mV) is in a good agreement with the data obtained at 77 K.

Stability of the obtained structures is another subject for a discussion. Nanoparticles were fixed in gaps only with weak van der Waals interaction of octanethiols with the substrate and/or the electrodes of a transistor. Despite that the most transistors have been shown a single-electron behavior during several days. Moreover they had been several times cooled to 77 K and warmed to 300 K. That fact shows that the nanoparticles fixation was sufficiently reliable. Looking forward, dithioles often used for nanoparticles fixation on a surface must significantly increase reliability of the system [14]. It shows a prospects of their use for creating of single-electron devices.
4. RESULTS

Molecular single-electron transistors based on small (2 - 4 nm) gold nanoparticles were fabricated using an electron-beam lithography and the electromigration method. Electrical characteristics of the obtained transistors were measured at 77 K and 300 K. The characteristics show that the regime of correlated tunneling of electrons was realized at 77 K. Single-electron effects are also noticeable at 300 K despite of the high level of thermal fluctuations that strongly reduces their clearness. Dithioles using for nanoparticles fixation will significantly increase reliability of the system and will create a real perspective for creation of practically useful single-electron systems based on nanoparticles.

This work was supported by "Russian fond of basic research" (grants № 12-07-00816-a, № 14-07-31328)

REFERENCES