# Fabrication of metallic electrodes with nanometer separation by electromigration

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A simple yet highly reproducible method to fabricate metallic electrodes with nanometer separation is presented. The fabrication is achieved by passing a large electrical current through a gold nanowire defined by electron-beam lithography and shadow evaporation. The current flow causes the electromigration of gold atoms and the eventual breakage of the nanowire. The breaking process yields two stable metallic electrodes separated by ~1 nm with high efficiency. These electrodes are ideally suited for electron-transport studies of chemically synthesized nanostructures, and their utility is demonstrated here by fabricating single-electron transistors based on colloidal cadmium selenide nanocrystals. © *1999 American Institute of Physics.* [S0003-6951(99)03427-0]

Electrical characterization of individual molecules[1–3] and nanocrystals[4–6] has received considerable attention recently in the scientific and engineering communities. Electron transport through these chemical nanostructures is strongly affected by electron–electron repulsion and energy-level quantization, and transport experiments can provide detailed insight into the electron dynamics.[7] The ability to make electrical devices based on individual molecules and nanocrystals is technologically relevant as well because it allows the fabrication of new types of devices whose function is defined by the chemical identity of the components.[8,9]

Despite considerable progress during the last decade, however, the field of nanometer-scale electron transport is still in its infancy. One of the major obstacles impeding further progress is the lack of reliable methods to interface a chemically synthesized nanostructure to macroscopic electronic circuits. In its simplest realization, an electrical measurement requires the fabrication of metallic electrodes whose separation is comparable to the size of the nanostructure itself. The resolution limit of current lithographic techniques remains, however, on the order of 10 nm, and consequently, the reproducible generation of the required electrodes is difficult by conventional fabrication techniques.

Recently, many different approaches have been put forward to address this problem.[1–6,10–20] They can be broadly divided into two categories, one that employs scanning tunneling microscopy (STM),[1,3,4,10,20] and the other that aims at defining a nanometer-sized tunnel gap through fabrication methods.[2,5,6,11–19] Both types of approaches have yielded fruitful scientific results. However, the conventional STM approach suffers from the lack of mechanical and electrical flexibility, and most fabrication methods reported so far exhibit low yields.

In this letter, a simple yet highly reproducible method to generate two metallic electrodes with a nanometer-sized gap is presented. The fabrication is based on the breakage of metallic nanowires using electromigration of metal atoms. Electromigration, which refers to the atomic motion in a conductor subject to large current density, has long been known to be the major failure mode in microelectronic circuitry.[21] Here, this failure mode is exploited advantageously to break gold nanowires in a controllable and self-limiting fashion. The breaking process consistently produces two metallic electrodes whose typical separation is about 1 nm. These electrodes can be readily adapted for electron-transport studies of molecules and nanocrystals, as demonstrated here by the transport measurements of individual cadmium selenide (CdSe) nanocrystals.

The electrode fabrication process starts with the generation of gold nanowires using conventional electron-beam lithography and shadow evaporation. This step has been described in detail previously,[5,12] and only the modifications pertinent to the nanowire fabrication are presented here. Electron-beam lithography on a PMMA/P(MMA–MAA) bilayer resist was employed to create a 200-nm-wide resist bridge suspended 400 nm above a $SiO_2$ substrate. Metallic nanowires were generated by evaporating 35-Å-thick chromium and 100-Å-thick gold at $\pm15°$ angles with respect to substrate normal. These fabrication parameters are chosen such that two electrodes generated by successive angle evaporation steps overlap to form a metallic wire. A representative scanning electron microscopic (SEM) image of a fabricated nanowire is presented in Fig. 1(a). As will be shown below, the overlapping region serves as the locus where a nanometer-sized tunnel gap is formed. Finally, 35-Å-thick chromium and 800-Å-thick gold were deposited straight down through the resist bridge to ensure a reliable bonding between nanowires and gold bonding pads defined by optical lithography.

The formation of a nanometer-sized gap is achieved by breaking fabricated nanowires through a controlled passage of current. For the characterization of the breaking process, the voltage drop across a nanowire was monitored in a four-
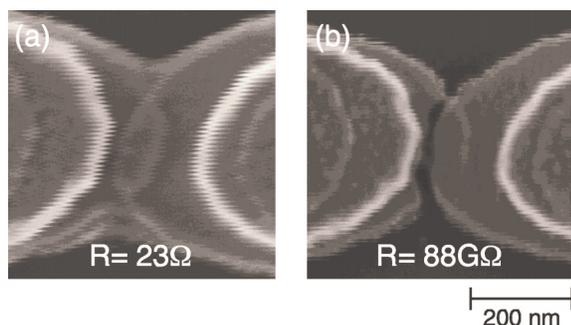
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FIG. 1. Field-emission scanning electron micrographs of a representative gold nanowire (a) before and (b) after the breaking procedure. The nanowire consists of thin (~10 nm) and thick (~90 nm) gold regions. In the images, diffuse white lines separate these two regions.
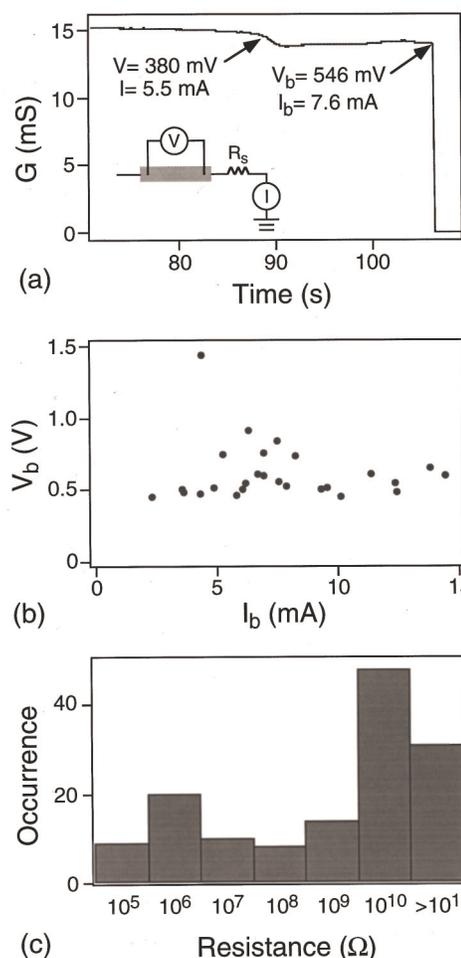


FIG. 2. (a) Representative conductance trace obtained during a nanowire-breaking procedure. The conductance is measured in a four-probe configuration schematically shown in the inset. The nanowire is broken by ramping a bias voltage through a 100 $\Omega$ series resistor at a rate of 30 mV/s. (b) Plot of current ($I_b$) and voltage ($V_b$) at the point of nanowire failure obtained from 26 nanowires prepared from five independent fabrication runs. All the nanowires were broken by ramping the bias voltage through a series resistor. (c) A histogram of the final tunnel resistances for 140 pairs of gold electrodes fabricated by nanowire breaking. Resistance values in each decade are collected as one bin.

probe configuration. Figure 2(a) shows a representative conductance trace during the breaking process. The conductance of a typical nanowire is on the order of 10 mS before the passage of current, and it remains constant at a small current level. As the current increases and the voltage drop reaches 300–500 mV, however, the conductance starts to change, indicating the onset of electromigration. Upon further increase of current, the conductance drops abruptly to almost zero, and the nanowire breaks. The whole process can be reproducibly performed as long as the thickness of a gold nanowire is below 20 nm. A representative SEM image of a broken wire is shown in Fig. 1(b). Inspection of many SEM images reveals that the failure of a nanowire typically occurs near the region where two angle-evaporated electrodes overlap, presumably because of the high resistance associated with the region.

Figure 2(b) shows a plot of voltages and currents at the nanowire break point obtained from 26 nanowires prepared from five independent fabrication runs. For all the data shown in Fig. 2(b), voltage biasing through a series resistor was employed to break nanowires, although different methods of raising current were also found to achieve essentially the same results. As clearly illustrated in Fig. 2(b), nanowire failure occurs consistently when the voltage drop across a wire exceeds a threshold of 470 mV. On the other hand, experimental parameters such as current at the breaking point, the total energy, and the average power dissipated during the breaking process (typically, on the order of $1 \mu J$ and 10 nW, respectively) were found to vary significantly from nanowire to nanowire. The latter observation suggests that the Joule heating of the junction is not the major mechanism behind the nanowire breakage, although the increase in temperature may affect the process indirectly.

According to the conventional understanding of electromigration, the motion of atoms subject to large current density arises from the direct action of an external electric field on a charged defect (direct force) and the momentum transfer from the conduction electrons to a defect (electron-wind force).[21–23] The direct force is proportional to the electric-field strength at the defect site, whereas the electron-wind force is proportional to the local current density. Inside a metallic nanowire both forces are expected to increase with the voltage drop because the current density is directly proportional to this voltage.[22,23] Within this framework, the observation of a threshold break voltage indicates the existence

of a force (or energy) threshold required to induce electromigration of gold atoms. Unfortunately, the quantitative determination of the force threshold requires a knowledge of the voltage distribution inside a nanowire, and hence, is not possible based solely on data presented here.

As described previously, electromigration of metal atoms has long been known to be the major failure mode of narrow conduction channels,[21] and therefore, the observation of nanowire breakage due to large current flow is not surprising in itself. A remarkable feature of the breaking process is, however, that it consistently yields two metallic electrodes with a measurable tunnel resistance between them. Figure 2(c) shows the histogram of tunnel resistances measured from 140 pairs of electrodes fabricated using this method. The broken wires typically exhibit low-bias tunnel resistances around 10 G$\Omega$. In some instances, resistance values as small as a few hundred k$\Omega$ were observed, whereas for about 15% of broken nanowires resistances larger than 200 G$\Omega$ were seen. Resistances larger than 200 G$\Omega$ could not be
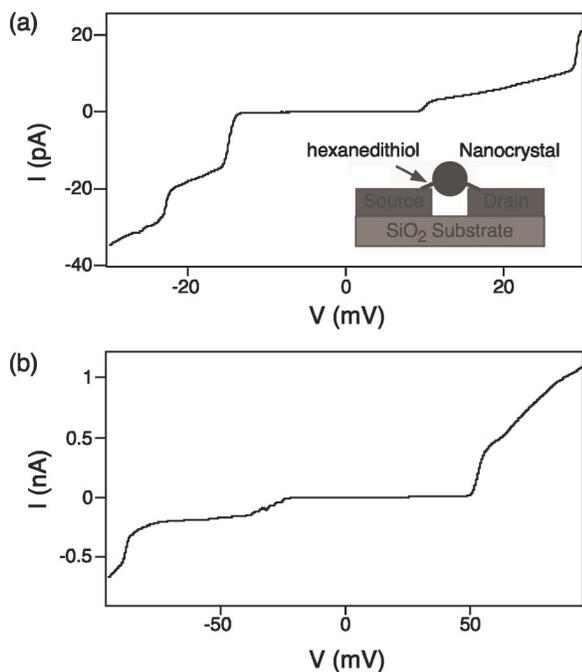
FIG. 3. Current vs voltage curves at $T = 1.5$ K obtained from single-electron transistors based on (a) 5 nm CdSe nanocrystals and (b) 7 nm CdSe nanocrystals. The inset in (a) shows the schematic diagram of an idealized device.

measured accurately due to current leakage through the $SiO_2$ substrate.

Previous tunneling measurements have shown that the tunnel resistance between two gold electrodes separated by $\sim 1$ nm is on the order of $G\Omega$ and that it increases exponentially as a function of distance with a characteristic length of $\sim 1$ Å.[11,18] Hence, the resistance values measured from broken nanowires indicate that the separation between two electrodes ranges from a few Å to $>1$ nm with a median of $\sim 1$ nm. The SEM images of broken nanowires, such as that shown in Fig. 1(b), also reveal that two electrodes are separated by less than 5 nm (roughly the resolution limit of our instrument), supporting the conclusion drawn from tunnel resistance measurements.

These electromigration-fabricated electrodes are ideally suited for electron-transport studies of molecules and colloidal nanocrystals. Indeed, these electrodes were successfully employed to make single-electron transistors based on individual CdSe nanocrystals. The CdSe nanocrystals were made by a colloidal synthetic technique,[24] and they were placed between two electrodes by the solution-phase deposition following the formation of self-assembled monolayers of 1,6-hexanedithiol on gold electrodes.[5,12] The linear dithiol molecules act as a bifunctional linker between nanocrystals and gold electrodes and also as a tunnel barrier between them. The nanowire-breaking process can be performed either before or after the nanocrystal deposition process because nanocrystals do not act as a major conduction path. The junction formation after the nanocrystal deposition can be advantageous because it allows the *in situ* generation of a single-electron transistor inside a measurement apparatus.

Upon the nanocrystal deposition, up to 60% of broken wires showed single-electron tunneling characteristics, depending on nanocrystal coverage. Figure 3 shows current versus voltage ($I-V$) curves at $T = 1.5$ K obtained from two representative single-electron transistors fabricated using this method. Both $I-V$ curves in Fig. 3 clearly exhibit the suppression of electrical current at low-bias voltages, known as the Coulomb blockade, characteristic of single-electron tunneling through nanometer-sized structures.[7] It was also found that the width of these Coulomb blockade regions could be tuned by applying the gate voltage that changes the electrostatic potential of the nanocrystal. Detailed electrical measurements of these CdSe-nanocrystal single-electron transistors will be discussed in a future publication.[25]

To conclude, a simple and highly reproducible method based on electromigration is developed to fabricate metallic electrodes with a typical separation of $\sim 1$ nm. The fabricated electrodes are stable in various experimental conditions including varying temperature and magnetic field, and are ideally suited for electron-transport measurements of chemically synthesized nanostructures. Their utility was demonstrated by the fabrication of single-electron transistors based on colloidal CdSe nanocrystals.

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