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Fabrication of reproducible sub-5 nm nanogaps by a focused ion beam and observation of Fowler-Nordheim tunneling

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Creating a stable high resistance sub-5 nm nanogap in between conductive electrodes is one of the major challenges in the device fabrication of nano-objects. Gap-sizes of 20 nm and above can be fabricated reproducibly by the precise focusing of the ion beam and careful milling of the metallic lines. Here, by tuning ion dosages starting from 4.6×10^{10} ions/cm and above, reproducible nanogaps with sub-5 nm sizes are milled with focused ion beam. The resistance as a function of gap dimension shows an exponential behavior, and Fowler-Nordheim tunneling effect was observed in nanoelectrodes with sub-5 nm nanogaps. The application of Simmon's model to the milled nanogaps and the electrical analysis indicates that the minimum nanogap size approaches to 2.3 nm. © 2015 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4930821>]

Research efforts in the fabrication of nano-sized devices have significantly increased in the recent years, and the fabrication of nanogaps between electrodes plays the key role in the building nanoparticle, nanocluster, and single molecules based devices.^{1–4} Direct writing methods, such as electron beam lithography (EBL) and other similar lithography techniques, are widely used in the fabrication process of the nanogaps with the gap size larger than 20 nm.⁵ However, approaching sub-10 nm gaps with high yields is not possible using EBL due to the resolution limit of standard electron beam writing, pattern development, and the metal lift-off process.⁶ High precision during gap fabrication with a 2–3 nm spacing can be achieved with special techniques such as mechanically controlled break-junctions, though their applicability to date is limited to the laboratory scale.⁷ For electromigration gaps^{8–10} that are usually fabricated by passing current through thin metal wires deposited on insulating substrates, it is difficult to control the gap size precisely and the fabrication yield of sub-10 nm nanogaps is only 50%.^{11,12} Furthermore, the electromigration process will depend on the local morphology of the grains that constitute the electrode material. This fundamentally limits the reproducibility of the gap with electromigration technique.

Focused ion beam (FIB) by milling thin metallic wires has shown remarkable advantages and progress in the fabrication of nanogaps.^{13,14} FIB milling is a maskless technique and widely employed to mill reproducible structures down to nanometer precision. The combination of EBL, photolithography, and FIB, where each technique is used at the length scale where it is best performing, makes FIB based nanogap fabrication an efficient way to produce a large number of nanogaps.^{4,15,16} Due to the reproducibility and repeatability in the fabrication along with the physical properties, FIB milled nanogaps are effectively employed in the analysis of

physical properties of single or few nano-objects such as nanoparticles and molecules.^{3,9,10} Reproducible nanogaps fabricated by FIB milling are more than 10 nm wide due to the limitation of ion beam diameter and profile, and, up to now, there is no systematic study on how to reach sub-5 nm nanogaps by using FIB milling.

In this paper, through precise controlling of the applied ion dosages, a total of 48 cut nanogaps were fabricated with the sizes from sub-5 nm to 30 nm. For a 250 nm wide gold wire (60 nm in thickness with 3 nm thick titanium as adhesion layer), an ion dosage of 5×10^{10} ions/cm can be utilized to fabricate sub-5 nm nanogap. Fowler-Nordheim tunneling effect was observed in the FIB milling sub-5 nm nanogaps, and the Simmon's model was applied to obtain the gap size.

The 250 nm wide wires were patterned on 300 nm thick SiO₂ on silicon substrate using EBL (Nanometer Pattern Generation System). The wire contains an evaporated 3 nm of titanium as adhesion layer along with a 60 nm thick gold layer (Fig. 1(a)). The gold wire was connected to the large contact pads by standard photolithography techniques.^{4,17} The nanogaps were milled into the gold wires with a focused Ga⁺ ion beam (30 keV in FEI Strata DB235). The precise focusing of Gallium ion beam at 1 pA enabled a probe size of about 7 nm. Series of nanogaps in gold wires were milled with ion dosages from 4.6×10^{10} ions/cm to 9.2×10^{10} ions/cm. The resistances of nanogaps were measured in a probe station (Agilent B1500), and the noise level of the current measurement was lower than 50 A. The applied bias voltages are limited between ± 0.6 V to avoid the high electrical field that could modify or destroy the nanogaps. To avoid the leakage current, current flowing through SiO₂ barrier to Si substrate and flowing back through SiO₂ to the current collecting electrode, we maintained the potential of the Si substrate at the same potential as the current collecting electrode (i.e., ground). Thus, the leakage current will not contribute to measured current. Furthermore, all the electrodes were cleaned for 5 min in an UV-ozone photoreactor (PR-100, UVP, Inc.) prior to the

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FIG. 1. (a) SEM image of 250 nm wide gold wire. SEM image of gold wires after FIB milling, under ion dosages of 4.6×10^{10} ions/cm (b), 5×10^{10} ions/cm (c), 6.7×10^{10} ions/cm (d), 8.3×10^{10} ions/cm (e). To have a clear view of (c), the sample under ion dosage of 5×10^{10} ions/cm is tilted at 55° shown in (f).

electrical measurements. Scanning electron microscopy (SEM) micrographs were recorded using a Zeiss Merlin SEM to determine the size of nanogaps.

Figs. 1(b)–1(e) show the typical nanogap evolution with increasing ion dosage. At low ion dosage (4.6×10^{10} ions/cm) as shown in Fig. 1(b), the sputtering of gold is not sufficient and the nanogap is not completely milled. With the increase in ion dosage of focused ion beam, the SEM images suggest that the nanogap starts to be fully formed from an ion dosage of 5×10^{10} ions/cm on Fig. 1(c). With increasing ion dosage, the gap width increases. The tilted view of Fig. 1(c) shown in Fig. 1(f) gives more insight in the details of the structure of the Au side walls of the gap where the tapering sidewall is observed, i.e., some parts of the nanogap sidewall reach more, some less into the nanogap. In fact, Unocic *et al.*¹⁸ reported differential sputtering of grain boundaries versus matrix using Ga ions in the FIB. In the nanogaps sputtered here, this differential sputter effect causes grain boundaries to be sputtered more compared with gold matrix. In turn, this effect creates nanometer sized protrusions reaching into the gap. Thus, the nanogap width at these protrusions becomes smaller. From the SEM images, we observe these protrusions caused by the FIB sputtering process on the bottom of the groove where the nanogap reaches its smallest distance. From the SEM image, we can estimate smallest Au electrode distance in this nanogap to be smaller than 5 nm though the accurate gap size cannot be measured from the SEM images.

FIG. 2. (a) The resistance as a function of gap dimension with error bars. The resistance is determined by linear curve fitting of the I - V characteristic, and the gap dimension is obtained from the SEM observations. (b) Plotted curve of $\ln(R)$ versus gap sizes with error bars.

Resistance measurements from the *current-voltage* (I - V) characteristic confirm the nanogap formation for ion dosages larger than 5×10^{10} ions/cm (Fig. 2(a) inset) with resistance of the nanogap above 12 TΩ. Whereas the nanogap width can be measured from SEM images with high accuracy for the gap sizes above 10 nm, for the sub-5 nm nanogap, the measurement from the SEM images is not accurate enough and we use electrical measurements to obtain the gap width. When the gap is fabricated, the two metallic electrodes are separated, introducing an energy barrier that leads to a tunneling current usually shows a linear behavior as a function of applied bias. Fig. 2(a) indicates the plot of the average resistance as a function of average gap size with standard deviation as an error, which displays a typical exponential curve. Inserted graphs are I - V characteristics of different nanoelectrodes. Through this plot, it is seen that all the nanogaps have resistances that are high enough to measure current through even single conductive molecule, i.e., above tens of TΩ. The exponential increase of the resistivity with the gap size reflects the transport being dominated by tunneling effect being expressed as the equation¹⁹

$$R = k \times \exp\left(\frac{2\sqrt{2m\phi}}{\hbar}D\right), \quad (1)$$

where k is a constant, ϕ is the barrier height, m is the electron mass, D is the nanogap size, and $\hbar$ is the reduced Planck

FIG. 3. *IV* characteristic of sub-5 nm nanogap (red dotted line) as well as a fit of Simmon's model (black solid line) with the accuracy of 5%. The fitting parameters are 2.3 nm for gap size and 0.8 eV for barrier height. Inserted are the Fowler-Nordheim representation plotted as $\text{Ln}(I/V^2)$ vs $1/V$.

constant. By plotting $\text{Ln}(R)$ as a function of D (Fig. 2(b)), the barrier height can be extracted and is estimated as 1 eV.

In large nanogaps (larger than 5 nm), Fowler-Nordheim effect is not easy, whereas for electrodes with sub-5 nm nanogaps, it is possible to observe the Fowler-Nordheim tunneling effect in the bias voltage range where the Au-electrodes are stable. The current flowing through the nanogaps at the applied bias voltages is a result of tunneling of electrons between the two gold electrodes. During the current-voltage measurements on the sub-5 nm nanogaps (Fig. 3 dotted line), direct tunneling occurs at the low applied bias voltage regime before Fowler-Nordheim tunneling, while the bias is high than the threshold voltage of Fowler-Nordheim, the current increases exponentially with the bias voltage. The Simmon's model is used to obtain further evidence for the gap size.^{20,21} The current density relationship for a generalized barrier can be expressed as below

$$J = \frac{4\pi me}{\hbar^3} \int_0^E D(E_x) dE_x \int_0^\infty f(E) - f(E + eV) dE_r, \quad (2)$$

where $D(E_x)$ is the tunnel probability of an electron with a kinetic energy of E_x . The tunnel probability can be obtained by the WKB approximation

$$D(E_x) \approx \exp\left[-A(\eta + \bar{\varphi} - E_x)^{\frac{1}{2}}\right], \quad (3)$$

where η is the Fermi level, $\bar{\varphi}$ is the mean barrier height given as $\bar{\varphi} = \frac{1}{D} \int_{x_1}^{x_2} \varphi(x) dx$, and $A = \frac{4\pi\beta D}{\hbar} (2m)^{\frac{1}{2}}$. If we define $D = x_2 - x_1$ as the gap size, then the current density equation can be expressed as below

$$J = \left(\frac{e}{2\pi\hbar D^2}\right) \left\{ \left(\varphi - \frac{eV}{2}\right) \exp\left[-\frac{4\pi D}{\hbar} (2m)^{\frac{1}{2}} \left(\varphi - \frac{eV}{2}\right)^{\frac{1}{2}}\right] - \left(\varphi + \frac{eV}{2}\right) \exp\left[-\frac{4\pi D}{\hbar} (2m)^{\frac{1}{2}} \left(\varphi + \frac{eV}{2}\right)^{\frac{1}{2}}\right] \right\}. \quad (4)$$

In this equation, the barrier height and the effective gap size act as parameters and can be fitted from the measured *IV* characteristics. The solid line shown in Fig. 3 is the fitted curve, while the dotted line is the experimental data. The barrier height is obtained as 0.8 eV, which agrees well with the estimated value and is consistent with reported work.^{21,22} Fitted effective gap size in this electrode is 2.3 nm with the accuracy of 5%, that is corresponding to the smallest distance between protrusions in Fig. 1(f). To ensure this result, five different electrodes are fabricated using the same conditions. Each nanogap shows similar *IV* characteristics with an average width of $2.6 \text{ nm} \pm 0.4 \text{ nm}$, which means that this process is highly repeatable. When we display the *IV*-data in a $\text{Ln}(I/V^2)$ vs $1/V$ plot, the curve shows a typical signature of Fowler-Nordheim tunneling.

In conclusion, by precisely tuning the ion dosage in FIB milling, we could achieve gold nanogap width down to 2.3 nm at an ion dosage of 5×10^{10} ions/cm and vary the gold nanogap width obtain a scaling relation for the gap size as a function of ion dosage. All nanogaps are insulating with resistance varying from tens of TΩ to thousands of TΩ corresponding to the gap sizes. Fowler-Nordheim tunneling is observed in FIB milling nanogap of 2.3 nm. To realize a precise measurement of nanogaps, Simmon's model is introduced and the fitted barrier height for gold electrode is obtained as 0.8 eV, which is consistent with reported work. Due to the high reproducibility, the sub-5 nm nanogaps have the potential to be widely used in nano-objects, even single molecule devices.

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