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Clean Electromigrated Nanogaps Imaged by Transmission Electron Microscopy

Douglas R. Strachan,*,†,‡ Deirdre E. Smith,† Michael D. Fischbein,† Danvers E. Johnston,† Beth S. Guiton,‡ Marija Drndić,† Dawn A. Bonnell,‡ and Alan T. Johnson, Jr.*,†

Department of Physics and Astronomy and Department of Materials Science and Engineering, University of Pennsylvania, Philadelphia, Pennsylvania 19104

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ABSTRACT

Electromigrated nanogaps have shown great promise for use in molecular scale electronics. We have fabricated nanogaps on free-standing transparent SiN_x membranes which permit the use of transmission electron microscopy (TEM) to image the gaps. The electrodes are formed by extending a recently developed controlled electromigration procedure and yield a nanogap with approximately 5 nm separation clear of any apparent debris. The gaps are stable, on the order of hours as measured by TEM, but over time (months) relax to about 20 nm separation determined by the surface energy of the Au electrodes. A major benefit of electromigrated nanogaps on SiN_x membranes is that the junction pinches in away from residual metal left from the Au deposition which could act as a parasitic conductance path. This work has implications to the design of clean metallic electrodes for use in nanoscale devices where the precise geometry of the electrode is important.

Electromigrated nanogaps[1] have shown great promise as electrodes for use in molecular scale electronics.[2] A controlled electromigration procedure has recently been developed[3,4] which can be used to create a nanogap with conductance ranging from a few atoms channel to an opaque tunnel barrier.[3] Researchers have also used this controlled electromigration in the few-atom and tunneling regimes to investigate the magnetoresistance[5,6] of a quantum spin valve.[7] Though the well-anchored leads greatly reduce magnetostrictive effects in these junctions, the magnectoresistance is still expected to depend on the precise atomic-scale structure of the electrode.[6] Additional motivation for high-resolution imaging of nanogaps comes from recent reports of measured transport behavior reminiscent of the molecular devices on bare electromigrated electrodes, i.e., *without molecules*.[4,8] These measurements on bare electrodes likely reflect the transport characteristics through residual metallic particles that remain from the initial deposition or electromigration procedures. Since molecular device yields are typically quite low (∼10% or less), distinguishing between devices with and without residual Au particles is imperative. Unfortunately, scanning electron microscope (SEM) images of nanogaps do not have the ∼1 nm resolution required to image the detailed structure of the nanogap.

To address this issue, we fabricated electromigrated nanogaps on free-standing SiN_x membranes that are transparent to high-energy electrons, and thus are compatible with transmission electron microscopy (TEM) at nanometer resolution. TEM images demonstrate that nanogaps with ∼5 nm electrode separation are reproducibly obtained and that the gap region is clear of debris with well-defined interfaces, making them highly suitable for molecular scale devices. One factor contributing to the clean gap region is that the junction formed by electromigration pinches in away from residual metal left from the Au deposition which could act as a parasitic conductance path. The nanogaps are mechanically stable on the order of hours, as determined by TEM, but relax over time (months) by the surface energy of the Au electrodes.

To fabricate the electrodes, we start with ⟨100⟩ silicon wafers polished on both sides with a deposited 100 nm low-stress silicon nitride layer.[9] Photolithography followed by reactive ion etching in a SF_6 plasma is used to define ∼700 μm square openings in the SiN_x surface on one side of the wafer. The exposed Si is then anisotropically etched using an aqueous KOH solution, which leaves a suspended silicon nitride membrane (∼40 μm square). We then use electron-beam lithography and thermal evaporation to define 40 nm thick Au leads connected by a narrow 100 nm constriction. These leads differ from the ones typically used on bulk Si substrates in that they do not have a Cr adhesion layer.

After fabrication, we form the nanogap using a computer-controlled electromigration process.[3] We find that samples on SiN_x membranes are more sensitive to destruction from static discharge than those on bulk silicon wafers. To protect

* To whom correspondence may be addressed: Douglas R. Strachan, drstrach@sas.upenn.edu; Alan T. Johnson, Jr., cjohnson@physics.upenn.edu.
† Department of Physics and Astronomy.
‡ Department of Materials Science and Engineering.

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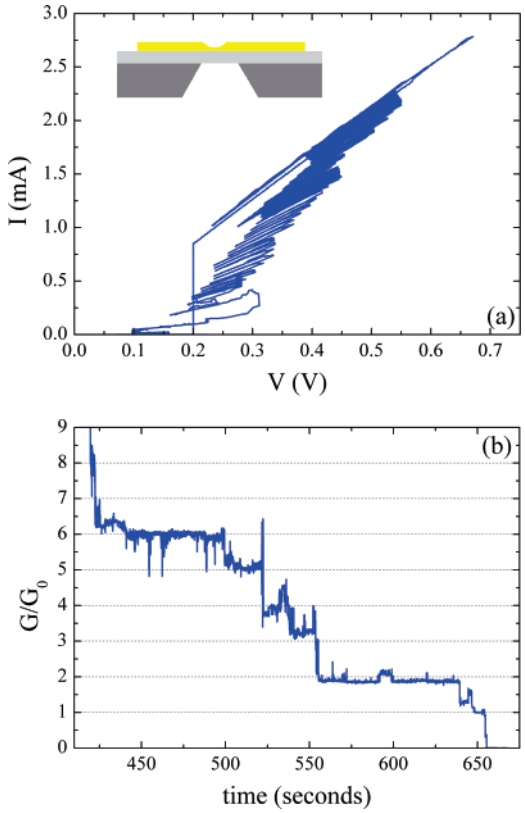

Figure 1. Current−voltage characteristics obtained during controlled electromigration of an Au wire on a 100 nm thick SiN_x membrane (see schematic inset). The electromigration procedure is stopped when the sample resistance reaches 1.5 kΩ. The sample voltage is held at 100 mV, and the sample conductance is monitored, giving the data shown in (b). (b) The time evolution of the conductance of this sample shows a series of jumps of order $G_0 = 2e^2/h$ in size.

the samples, we first connect the Au leads to a 100 kΩ potentiometer set to its maximum resistance. Figure 1a shows the series of current−voltage characteristics that are obtained during the controlled electromigration process.

The evolution of the junction through the first two stages (i.e., the bulk-neck and few-atom channel regimes) occurs similarly to the one performed on bulk substrates.[3] The voltage applied to the sample is increased at 40 mV/s, and the resistance is monitored. When the voltage reaches 0.2 V during the initial ramp, the potentiometer is zeroed so that the controlled electromigration can proceed. When a specified threshold for $\Delta R/R$ is reached (where R is defined at the start of a voltage ramp) the program reduces the voltage by approximately 100 mV at 400 mV/s. Compared to leads on bulk substrates, leads on SiN_x typically require threshold values half the size in order to prevent catastrophic failure.

To demonstrate that the electrode constriction can be narrowed down to the atomic scale on a membrane, the program is stopped when the nanogap resistance is roughly 1.5 kΩ and the voltage maintained at about 100 mV. As with wires on bulk substrates,[3] the wire slowly evolves under these conditions with abrup tjumps in conductance of approximately $G_0 = 2e^2/h$, demonstrating that the electromigration procedure yields an atomic scale ballistic conductor.

Although we find that controlled electromigration proceeds through the bulk-neck and few-atom channel regimes on SiN_x membranes as for bulk substrates, there is a marked deviation in the behavior of the final tunneling regime. On bulk substrates, the tunneling regime shows abrupt, approximately factor of 10 changes of the electrodes that are triggered by increasingly large voltage ramps.[3] This indicates atomic scale rearrangements of the electrodes due to the applied voltage.[10] In contrast, on SiN_x membranes when the conductance falls below G_0 the tunneling resistance quickly and uncontrollably evolves to greater than 10^{12} Ω (roughly the resolution of our instrumentation) on time scales of order seconds. This indicates that the nanogaps formed on SiN_x membranes are larger than those on bulk substrates.

Figure 2 shows a typical nanogap formed with controlled electromigration on a SiN_x membrane at various magnifications imaged with a JEOL 2010 TEM microscope under bright-field conditions (where Au appears dark). At low magnification (Figure 2a) the image shows the outline of the nanogap surrounded by small Au particles. These particles of Au, ranging in size from 1 to 10 nm, are formed during the Au evaporation and are present before and after the controlled electromigration procedure is performed. Imaging of these tiny particles illustrates the capability of detecting residual Au in the nanogap left from the electromigration. The images at high magnifications in parts b and c of Figure 2 show that the substrates are clear of such debris in the vicinity of the 5 nm nanogap, and thus the gaps appear free of parasitic conductance channels.

The inner boundary of the region in Figure 2a containing the Au particles roughly outlines the extent of the original leads before electromigration is performed. It is clear from this lower-resolution image that the controlled electromigration tends to pinch the junction in away from the edge of the original lead. We consistently see this effect in the controlled electromigrated nanogaps on SiN_x membranes, but it is not seen on bulk substrates. This difference could stem from a decreased temperature gradient in the vicinity of the forming nanogap due to weaker thermal sinking on the SiN_x membrane. The pinching effect on membranes has the significant advantage that the resulting nanogap is far away from the residual Au particles due to evaporation.

We find that controlled electromigration on SiN_x membranes consistently yields nanogaps of about 5 nm in size that are stable on the time scale of days. After fine tuning the parameters of the controlled electromigration, we obtain nanogaps with almost %100 yield. A typical assortment of these nanogaps is depicted in Figure 3, where both the course and fine features of five typical gaps are shown. Of a total of 17 nanogaps we have successfully made with controlled electromigration on SiN_x membranes and imaged with TEM, their average size is 5.9 ± 1.1 nm. The size of these nanogaps is roughly two to three times that of ones fabricated on bulk substrate, where gaps of about 2 nm in size are indicated by the tunneling resistance.[1,3] The larger gaps on SiN_x may be due to differences in the final shape of the electrodes formed on these membranes with those formed on bulk substrate.

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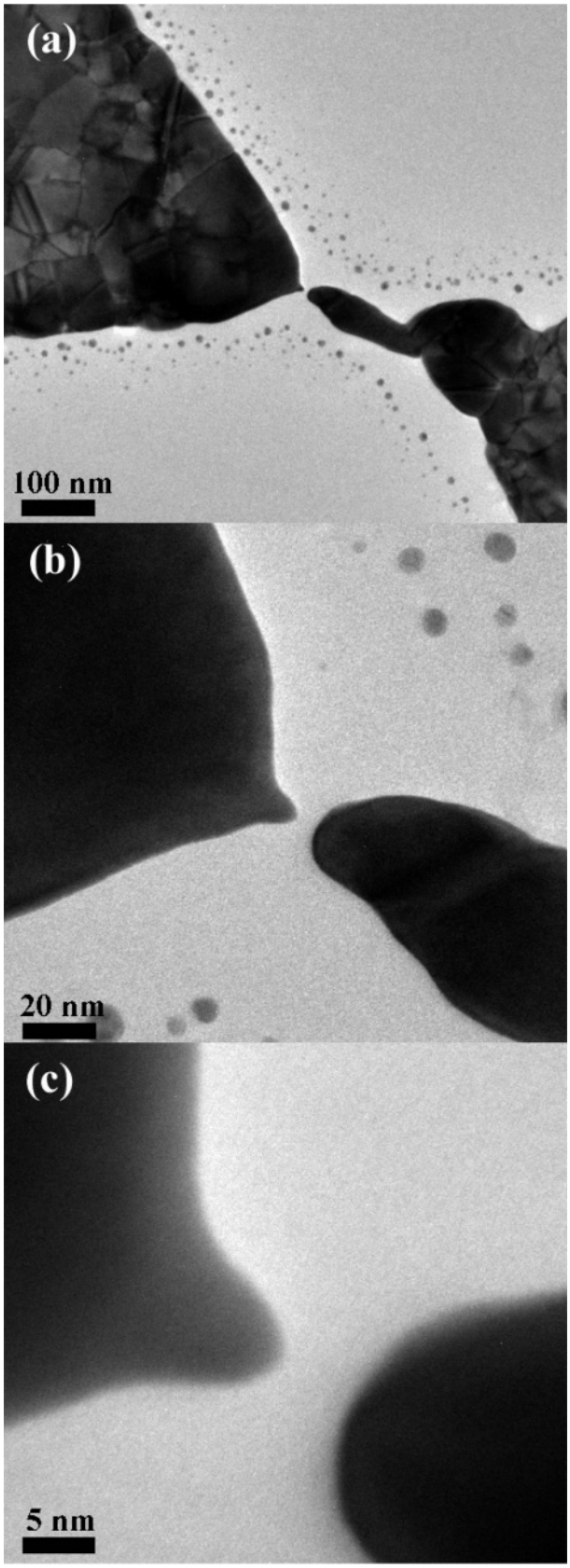

Figure 2. TEM images of a typical electromigrated nanogap on SiN_x membrane at three magnifications. The images were made approximately 1 day after the electromigration was performed.

Evidence that the shape of the electrodes has an influence on the stability of the nanogap is found by investigating their

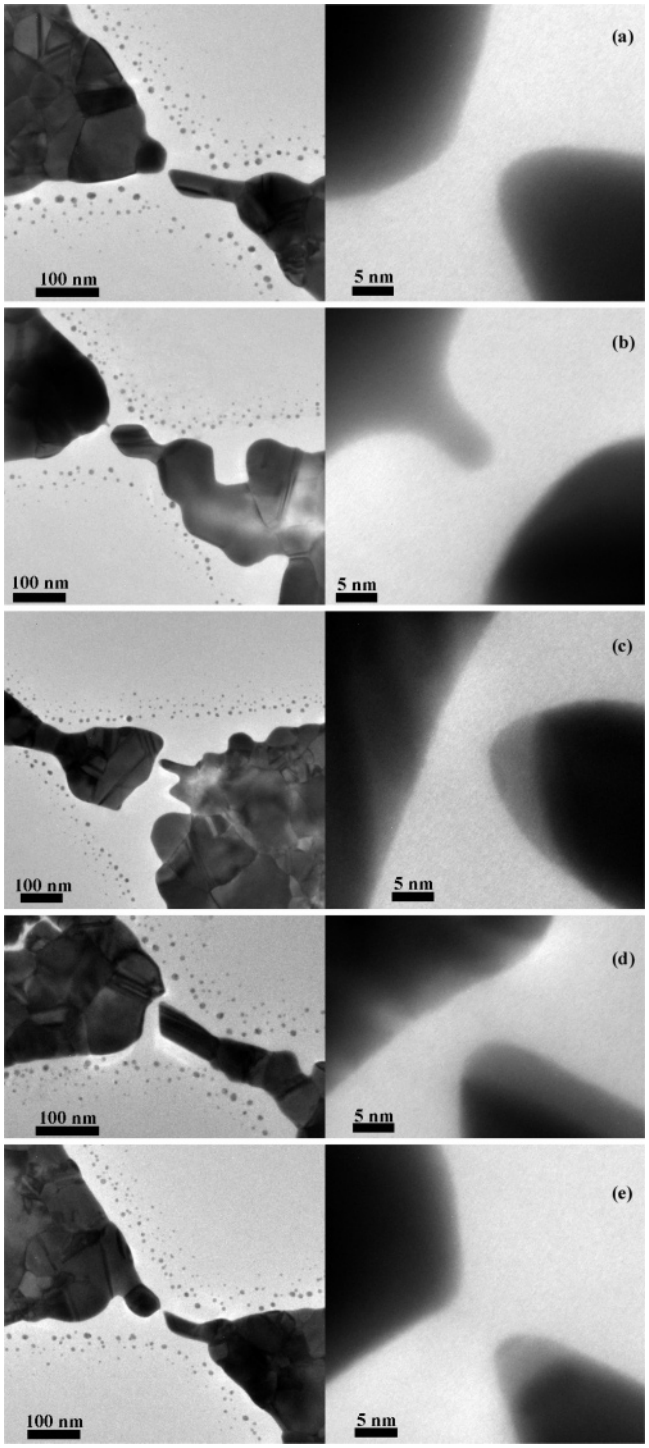

Figure 3. TEM images of an assortment of typical electromigrated nanogaps on SiN_x membranes. Each gap is imaged under both low and high magnification to illustrate the course and fine features of the resulting structures.

long-term evolution with TEM. Over the time span of a typical TEM imaging session (~1 h) there is no detectable evolution the nanogap size. Yet, if the nanogaps are left at room temperature for 3 months or more, then a slight recession of the gap is detectible. Figure 4 shows the same gap as in Figure 2 imaged after it had been left at room temperature for 4 months, with the initial contour represented by the dotted lines. The nanogap has clearly expanded, and

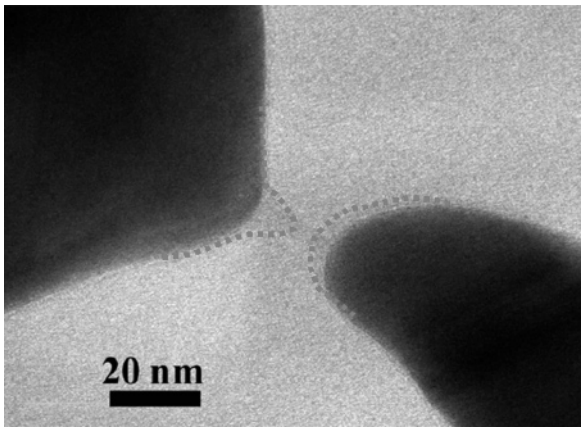

Figure 4. The same nanogap shown in Figure 2 after leaving the sample for 4 months at room temperature. The dotted lines represent the original contour of the leads approximately 1 day after the electromigration was performed.

the locations of greatest curvature have been smoothed out. This indicates that the gap evolves under the surface tension of the Au which tends to smooth out abrupt features. This evolution of the nanogap after electromigration is also consistent with recent reports of continued relaxation of Au nanogaps formed by electron bombardment even after the intense irradiation is completed.[11]

In conclusion, we have developed a controlled electromigration technique to fabricate nanogaps on free-standing transparent SiN_x membranes. Using TEM imaging, we determined that the electrodes are clear of residual debris that can be left from either the electromigration or evaporation procedures and that the 5 nm nanogaps are stable on the order of days. A major benefit of these electromigrated nanogaps on SiN_x membranes is that the junction pinches in away from the edges of the original leads. This avoids potential parasitic conductance channels through spurious metallic particles left from the metal evaporation. The slow evolution of the gaps on the 3 month time scale also suggests that the stable size of nanogaps is determined by relaxation of the surface energy of the Au electrodes. This work has implications for the design of clean metallic electrodes for use in nanoscale devices where the precise geometry of the electrode is important.

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References

(1) Park, H.; Lim, A. K. L.; Park, J.; Alivisatos, A. P.; McEuen, P. L. *Appl. Phys. Lett.* **1999**, *75*, 301.
(2) Liang, W.; Shores, M. P.; Bockrath, M.; Long, J. R.; Park, H. *Nature* **2002**, *417*, 725; Park, J.; Pasupathy, A. N.; Goldsmith, J. I.; Chang, C.; Yaish, Y.; Petta, J. R.; Rinkoski, M.; Sethna, J. P.; Abruna, H. D.; McEuen, P. L.; Ralph, D. C. *Nature* **2002**, *417*, 722.
(3) Strachan, D. R.; Smith, D. E.; Johnston, D. E.; Park, T.-H.; Therien, M. J.; Bonnell, D. A.; Johnson, A. T. *Appl. Phys. Lett.* **2005**, *86*, 043109.
(4) Houck, A. A.; Labaziewicz, J.; Chan, E. K.; Folk, J. A.; Chuang, I. L. *Nano Lett.* **2005**, *5*, 1685.
(5) Keane, Z. K.; Yu, L. H.; Natelson, D. cond-mat/0510094 2005.
(6) Bolotin, K. I.; Kuemmeth, F.; Pasupathy, A. N.; Ralph, D. C. cond-mat/0510410 2005.
(7) Chopra, H. D.; Sullivan, M. R.; Armstrong, J. N.; Hua, S. Z. *Nat. Mater.* **2005**, *4*, 832.
(8) Sordan, R.; Balasubramanian, K.; Burghard, M.; Kern, K. *Appl. Phys. Lett.* **2005**, *87*, 013106.
(9) Bean, K. E. *IEEE Trans. Electron Devices* **1978**, *ED-25*, 1185. Zhou, Y. X.; Johnson, A. T.; Hone, J.; Smith, W. F. *Nano Lett.* **2003**, *3*, 1371. Wafers were purchased from the U. C. Berkeley Microfabrication Laboratory (www-microlab.eecs.berkeley.edu).
(10) Huang, D.; Yamaguchi, F.; Yamamoto, Y. *Jpn. J. Appl. Phys.* **1998**, *37*, 3824; Trouwborst, M. L.; van der Molen, S. J.; van Wees, B. J. cond-mat/0510385 2005.
(11) Zandbergen, H. W.; van Duuren, R. J. H. A.; Alkemade, P. F. A.; Lientschnig, G.; Vasquez, O.; Dekker, C.; Tichelaar, F. D. *Nano Lett.* **2005**, *5*, 549.

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