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In-situ TEM observation of gold nanogaps by electromigration

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ABSTRACT

Electromigration is one of atom diffusion phenomena caused by electron flow. To apply this phenomenon on nanostructure fabrication, analysis of the atom transfer mechanism is very important. Based on this objective, in-situ transmission electron microscopy observation was performed on the narrowing process of polycrystalline Au wires. When the wire was composed of plural grains, the narrowing rate was relatively large. On the other hand, it was strongly suppressed when the wire width was less than the grain size. This indicates that the atom transfer along grain boundaries is the important factor for wire narrowing by electromigration. By continuous current flow, the wire was ruptured and nanogaps of about 10nm were fabricated. The trend of nanowire breaking (and the nanogap formation) was occurred in the vicinity of grain boundaries. The final stage of wire rupturing was analyzed in detail by in-situ TEM.

1. INTRODUCTION

In recent years, miniaturization of electronic devices has greatly advanced to integrate circuits for informatics. Responding to the needs of the times, various nano-scale fabrication techniques have been proposed, e.g. scanning probe microscopy (SPM) based lithography (Watanabe 2004) and shadow evaporation (Umeno 2009). The break junction process of metallic narrow patterns by using electromigration investigated in this work is one of such methods. As widely known, electromigration induces harmful effects which destructing integrated circuits. On the other hand, in a research field of nanofabrication, this effect is positively used to produce nanogaps and narrow contacts by controlling the current flow (Strachan 2008, Gao 2009). By using nanostructures fabricated by this method, there are several successful reports such as single electron transistor (SET) devices (Wolf 2010), memory devices (Naitoh 2008).

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For further development of this method as an established fabrication method in nanometer scale, changes in microstructure during the electromigration process should be investigated in more detail.

Based on these backgrounds, the rupture process of Au nanowires formed on the Si-N substrate was directly observed and analyzed by transmission electron microscopy (TEM) (Chen 2008, Murakami 2012). For dynamic observation during the electromigration process in nanoscale, in-situ TEM was used where the current measurements are simultaneously performed in the TEM instrument together with the observation of the structural change of the wire.

2. Experimental details

Device patterns were made by using photolithography, electron beam lithography and the lift-off process on Si-N /Si (100) substrate. First, electrode pads made of Au (35 nm)/Cr (5 nm) were prepared by using vacuum evaporation. Afterwards, wires (bridges) of 35 nm thick Au layer were deposited. Finally, window for TEM observation (approximately 100 x 100 μm) were formed from the substrate backside by using a KOH (25 wt. %) aqueous solution at 80°C. As a result, a 25- or 35-nm-thick self-standing Si-N membrane on which Au patterns to be observed are placed was obtained.

The sample was set in a custom-made double-tilt TEM holder with which the electric property can be measured through sixteen co-axial cables during the TEM observation. The TEM instrument used was a JEM-2010 (Jeol, C_s = 0.5 mm, vacuum ~ 10^{-5} Pa) microscope with a CCD video camera system. Electric measurements were performed in the constant voltage mode by using a Yokogawa GS610 source measure unit (SMU). The current was measured after adjusting the applied voltage manually.

3. Results and discussion

Nanowire on the final stage before rupture is shown in Fig. 1. Figure 1(a) was obtained by voltage application for about 3200 sec from the initial state (with manual control of 0 - 250 mV). The region with dark contrast corresponds to the gold wire. A series of images during further voltage application are presented as Figs. 1 (b) to (d). The triangle in each image denotes a grain boundary which was an indicator showing the corresponding position in Fig. 1(b) – (d). The voltage was 0.246 V, some of which may be consumed at other parts than the nanowire, and the current was between 3.84 and 3.74 mA. In these images, the electron flew from the right-top to the left-bottom. In Fig. 1 (a), smallest width of the gold wire was more than 50 nm. After 180 sec, the gold nanowire was elongated and became narrow (Fig. 1 (b)). Continuous current flow made the nanowire narrower and induced rearrangement and/or recrystallization of grains (Fig. 1 (c), (d)). The wire shape became more regular with crystal habit. Finally in Fig. 1 (d), the narrowest width of nearly 10 nm was formed at a grain boundary.

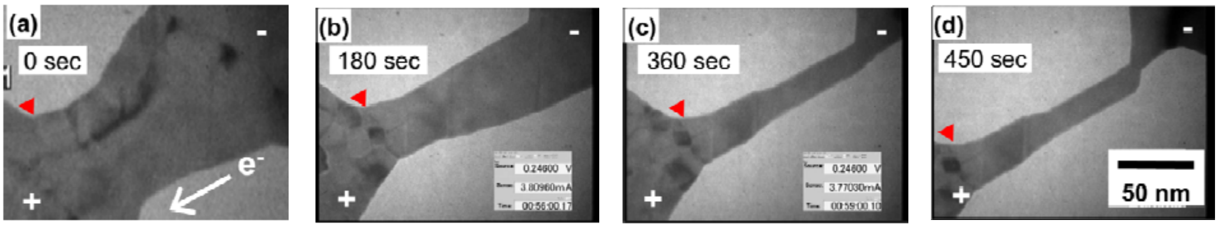

Fig. 1 TEM movie during narrowing process of an Au nanowire. Electrons flow from the right-top to the left-bottom as indicated by an arrow in (a). Plus and minus signs denote the voltage polarity. Positions indicated by red arrows correspond to each other.

To investigate whether the above mentioned phenomenon was surely caused by electromigration, the experiment with polarity alternation of voltage (i.e. direction alternation of electron flow) was performed. Electromigration may induce alternation of change in wire geometry, while such alternation should not occur by thermal effect. Typical images during the dynamical observation are summarized in Fig. 2 where triangles indicate a corresponding stacking fault existing in these TEM images. It should be noted that an arrow in each TEM image indicates the direction of electron stream which is opposite to the current flow. First, positive voltage was applied to the right-bottom side of the wire (Fig. 2 (a)) and thus the electrons flew from the left-top to the right-bottom. The narrowing occurred at the left side (i.e. upper side of the electron stream) (Fig. 2 (b)). After Fig. 2 (b), the voltage polarity was alternated, and thus the electron flow was inverted. In this case, areal reduction and gain was recognized at the upper and the lower sides of the electron stream (i.e. negative and positive voltage side), respectively (Fig. 2 (c)). This is clearly seen by comparing the image with the dotted line indicating the contrast edge of Fig. 2 (b). The position with areal reduction in Figs. 2 (b) and 2 (c) is inverted. Subsequent alternation was carried out between Figs. 2 (d) and 2 (e). The position with reduction and gain of area was again inverted. Continuing the voltage application, the wire became further narrow (Fig. 2 (f)). Narrowing occurred at the upper side of the electron stream, and widening at the lower stream side. This was satisfied for both voltage polarities. This result suggests that Au atoms were transported along the same direction as that of the electron flow. Thus, it is clearly proved that the phenomenon observed in this work was caused by electromigration. Unrelated to the voltage polarity, the wire became narrower.

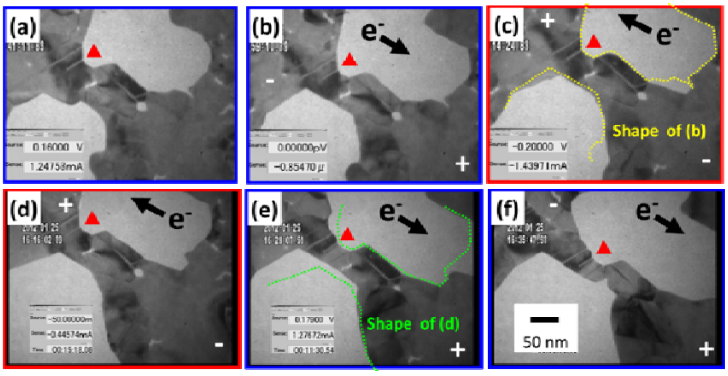

Fig. 2 Geometrical change during alternation of the current flow. Positions indicated by

red arrows correspond to each other. Plus and minus signs denote the voltage polarity. Arrows are the electron flow direction.

From series of observations, we confirmed that constricting and rupturing position was mainly around grain boundaries. A typical TEM image of a gold nanowire just before rupturing (i.e. formation of a nanogap) is shown in Fig. 3 (a). The wire width was around 5nm and the grain boundary was at the constriction. The electrons flew from the top to the bottom, thus atoms are thought to move along this direction. The wire width around the grain boundary oscillated randomly by time as shown in Figs. 3 (b)-(d). These figures show the time variation of nanowire width, and measuring positions correspond to the same colored arrows in Fig. 3 (a). On the other hand, the width change in the lower region was clearly different from and smaller than the upper region. It is expected that wire width change is influenced by an existing grain boundary. The atom movement beyond a grain boundary seems to be obstructed, and atoms restart to move in one breath with some trigger. Colored area in Fig. 3 (b) to (d) declares clear examples of these changes. Based on this assumption, the nanowire width is understood to oscillate randomly in the upstream region of the electron flow. In the downstream region, however, atoms may be more or less constantly transported, and wire width change has different trends from that in the upstream region. These results suggest the difference of atomic flow rate between up- and downstream regions separated by a grain boundary.

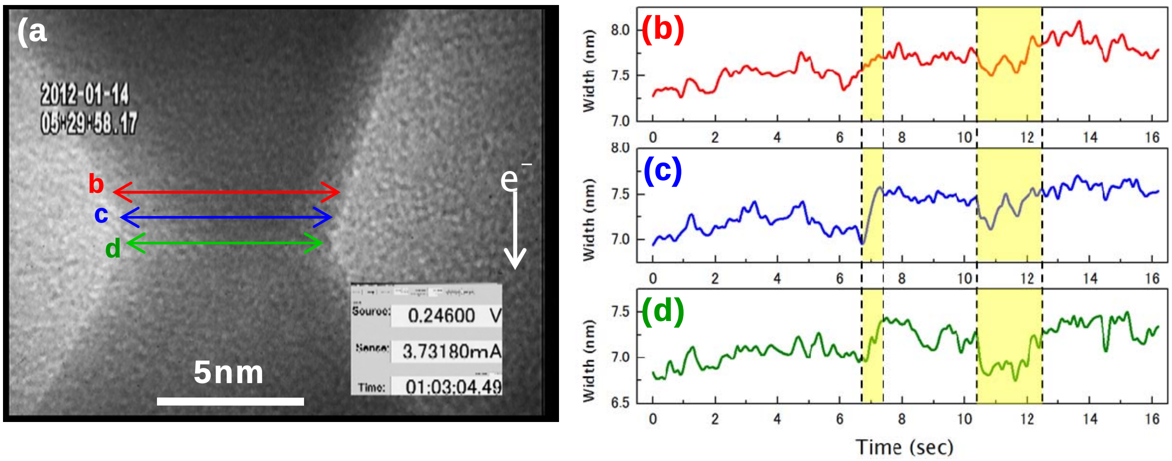

Fig. 3 (a) TEM image of a gold nanowire, and (b)-(d) time variation of the wire width.

By continuing the voltage application, the wire was ruptured and a nanogap was formed. An example is shown in Fig. 4. The dotted lines denote the grain boundaries. These positions were roughly decided by using lattice fringes observed in highly magnified images. In addition, solid curves indicating approximate positions of wire outlines are superposed. In Fig. 4 (a), the nanowire before rupture is shown. The nanowire width was approximately 3 nm. The nanowire was constricted at the vicinity of the grain boundary composed of three grains. Further elapse of 3.5 minutes, the constriction became narrower and its width diminished to approximately 1 nm as shown in Fig. 4 (b). At this stage, the contrast intensity is very weak because the thickness of the nanowire region became very thin. However, the electrical measurements indicated that the nanowire was still in connection.

Finally obtained nanogap is presented in Fig. 4 (c). The gap distance was approximately 11 nm though the constriction just before rupture was nm in size. Even after the electrical disconnection, the Au region around the gap moved like ameba, and the gap was expanded to about 10 nm. To produce well facetted extremely narrow gaps, well controlled current flow should be used at low temperature as proposed in earlier works. As a general tendency observed in this work, nanowire disconnection (and the nanogap formation) was occurred in vicinity of grain boundary.

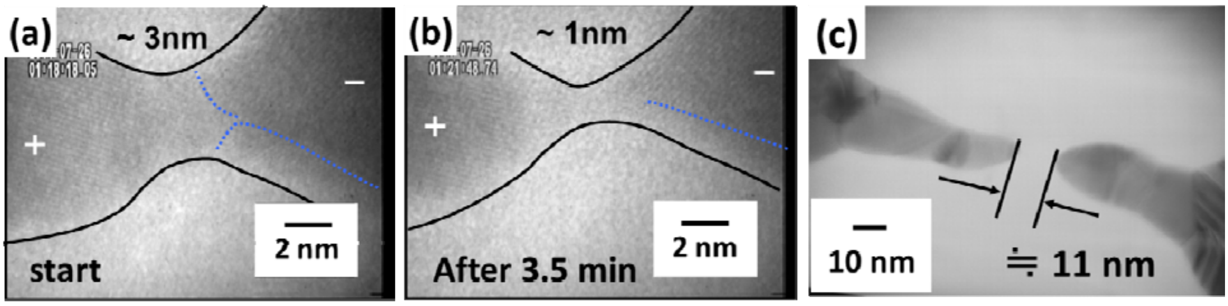

Fig. 4 (a)-(b): Images during the rupture process, and (c): fabricated nanogap.

3. CONCLUSIONS

In this study, the wire narrowing process by current flow was investigated about Au wire on Si-N by means of in-situ TEM. All of the results pointed out that the geometrical feature of the grain boundary plays an important role to control electromigration. First, we confirmed that the geometrical change and moving direction of Au atoms were along the electron stream. Thus, the observed phenomenon is concluded to be caused by electromigration. Second, we observed an effect of grain boundary on narrowing, which located perpendicularly to the wire axis. The grain boundary makes an imbalance of atom flow. The oscillating phenomenon observed here must relate to the origin that nanogaps tend to be formed around grain boundaries. Finally, rupturing process of Au nanowire was directly observed. The rupturing occurred at grain boundary and the distance of nanogap was around 10nm.

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