SUPPLEMENTARY INFORMATION

Optimization of the nanotwin-induced zigzag surface of copper by electromigration

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Supplementary materials:

Synthesis of copper film

A Cu film of 50 nm in thickness was deposited on the silicon nitride membrane by e-beam evaporation from 99.999%-pure source in a vacuum of 2×10^{-6} torr at room temperature. The deposition rate was approximately 1 Å/s. Then, the specimen was annealed at 250 °C for 1 min in a vacuum of 10^{-3} torr by a rapid thermal annealing system to stabilize the microstructure of the Cu line.

Experimental procedure

Thin Cu line specimens for *in-situ* TEM observations were prepared through conventional thin film deposition, photolithography and etching processes, as shown in Figure S1 (a). The specimen was mounted on a specially designed TEM holder that allows for introducing an electric current through the Cu line, as shown in Figure S1 (b). The sample was loaded into the TEM system (JEOL 2000V UHV-TEM) and inspected at an ultrahigh vacuum environment of 3×10^{-10} torr to prevent Cu oxide formation. *In-situ* TEM observations were performed when applying an electric current with an average density of 2×10^6 A/cm^2 through a 15 μm-wide Cu line specimen. We inspected several $(0\bar{1}1)$-oriented Cu grains with several types of twins. The current-driven TB behavior was video-recorded and analyzed.[1]

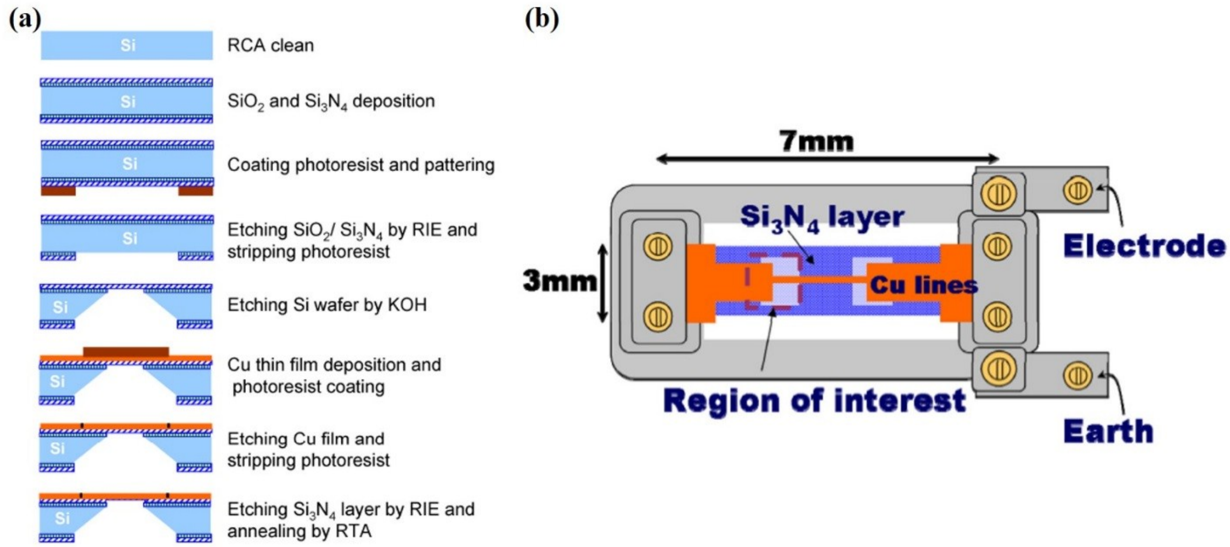

Figure S1. (a) Process sequences of preparing the free-standing Cu specimens for *in-situ* TEM observations. (b) Schematic diagram of the specially designed TEM holder with TEM sample.[1]

The EM induces a feedback selection of the $(\bar{1}11)$ /(200) zigzag pair when atomic layers are removed by step-edge migration. To demonstrate that the $(\bar{1}11)$ /(200) zigzag pair is indeed one of the EM preferred meta-stable surfaces, an alternative method was used to observe the surface evolution of a nanotwinned Cu by refilling a void (Fig. S2). The initial edge with a void is composed of facet {111} planes and stepped surfaces alternating in twin and matrix, as Fig. S2 (a) shows. During the EM-induced Cu refilling, the stacking sequence of incoming Cu atoms followed the pre-existed atomic symmetry, and the surface had grown to the $(\bar{1}11)$ /(111) zigzag pair due to its having the lowest surface energy. Due to the supplementation of Cu atoms by the EM flux, one of the facet $(\bar{1}11)$ surfaces gradually evolves into the stepped surfaces shown in Fig. S2 (c). Subsequently, the $(\bar{1}11)$ /(111) zigzag pair developed into the $(\bar{1}11)$ /(200)zigzag pair seen in Fig. S2 (d). The $(\bar{1}11)$ /(200)zigzag pair is preserved for up to 13 minutes without changes, and it is a dynamic equilibrium configuration according to Le Chatelier's principle.

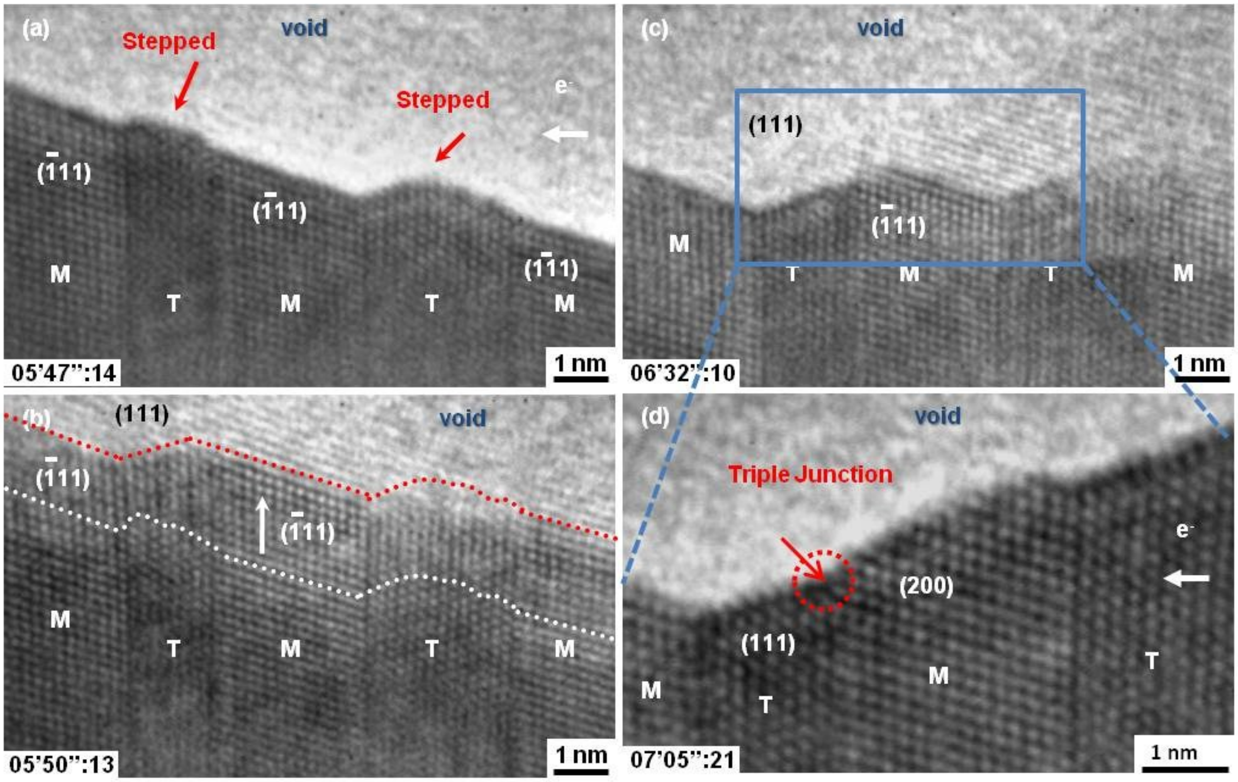

Figure S2. (a)-(d) HRTEM images of the$(0\bar{1}1)$-oriented Cu grain edge, revealing the EM-induced void refilling as a function of time. The first two numbers are in units of minutes, the second two numbers are in units of seconds and the following two smaller numbers are in units of 1/30 seconds. The dashed lines in (b) represent the original (white) and final (red) positions of the edge, and the upward arrow indicates the growth direction of the nanotwinned copper.

Supplementary movies:

Movie S1. An *in-situ* TEM-EM movie showing the real time movement of surface step-edges on the nanotwin-free region under current stressing. The movement shows the removal of {111} Cu atomic layers and changes in the surface morphology of Cu.

Movie S2. An additional *in-situ* TEM-EM movie showing the concurrent multiple step-edge migration in a twin-free $(0\bar{1}1)$-oriented Cu grain. The relative difference between the light and dark portions of the picture is contributed from the Cu thickness. The process of surface step-edge migration not only grows the void but also thins the Cu grain by removing {111} atomic layers.

Movie S3/S4. Two in situ movies showing the step-edge migration when a meta-stable zigzag surface is presented. The change of the surface morphology of Cu is subtle in the nanotwinned region compared to that in the nanotwin-free region, and the preserved zigzag surface relative to the nanotwin-free surface gives a statistically lower EM rate.

Reference

1. K. C. Chen, W. W. Wu, C. N. Liao, L. J. Chen and K. N. Tu, *Science*, 2008, **321**, 1066-1069.