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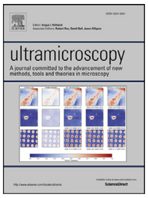

Preferred diffusion paths for copper electromigration by *in situ* transmission electron microscopy

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ABSTRACT

Ionic transport in the reverse direction of an electric field is caused by momentum transfer from free electrons to metal ions, *i.e.*, electromigration (EM), which is a critical factor leading to copper (Cu) interconnect failure in integrated circuits under extreme operating conditions. We investigated Cu self-diffusion paths under electrical bias using *in situ* transmission electron microscopy (TEM). An electric current was applied to multigrain Cu lines in the TEM instrument for durations of up to the order of 10^4 s to trace EM-induced Cu movement around voids and hillocks. Combining this approach with scanning nanobeam diffraction, we observed that high-angle grain boundaries exposed to the free surface are the most favored paths for Cu EM, rather than a specific orientation within the grain. On hillocks of accumulated Cu atoms, we directly observed grain growth, accompanied by the formation of $\Sigma7$ high-mobile and $\Sigma3$ twin coincidence site lattice boundaries for effective growth. This study provides insight into the EM mechanism to improve the reliability of metal interconnect design.

1. Introduction

Elemental diffusion is one of the fundamental processes involved in the physical phenomena of materials, such as phase transformation, sintering, and solidification. Diffusion can be enhanced or suppressed by external environmental variables imposed on the system, which can be effectively exploited in modern microelectronics to extend the lifetime expectancy of materials. As modern devices are pushed toward extreme operating conditions due to scaled-down design, electromigration (EM), *i.e.*, metal ion transport toward the positive electrode caused by momentum transfer from conducting electrons to metal ions, raises concerns regarding reliability. In particular, the EM phenomenon makes metal interconnect design one of the major constraints on circuit integration in downscaled devices, given that the gradual movement of metal ions in the interconnect may lead to void formation and result in eventual interconnect failure. Accordingly, there have been various studies on EM-induced self-diffusion paths in metal interconnects (particularly in copper (Cu), the most common material for interconnects), in an attempt to better understand the mechanism and extract appropriate design parameters for reliable interconnects [1–4]. Many such studies have been performed at the microscopic scale and under extreme experimental conditions [5–7]. With the aid of precision processing techniques and materials, it has become possible to fabricate a nanostructure suitable for making *in situ* observations of nanostructural changes influenced by external environment stimuli, such as electric current, heat, and mechanical stress. Dominant self-diffusion paths can be identified by observing mass transport behavior in Cu atoms using *in situ* transmission electron microscopy (TEM). One research group investigating Cu at the atomic scale discovered $<110>$-oriented surface diffusion as the preferential EM path; however, it is difficult to make generalizations based on this result, due to the limited observation area under study [8–9].

In this study, EM-induced self-diffusion paths in a Cu line were examined using *in situ* TEM to identify the failure mechanism in multigrain thin films. Applying scanning nanobeam electron diffraction (ASTAR, NanoMEGAS [10]), we constructed a crystallographic orientation map of nano-sized grains to the micro-scale, which allowed a comparison with previous findings reported at different scales. A comparison of the crystal orientation map and the mass-transport video recorded *in situ* revealed the most favored Cu migration paths for overall void expansion, which we identified as the surface-exposed high-angle grain boundaries as opposed to a specific grain orientation. In addition, we directly observed the grain growth process during hillock formation, which accompanied the formation of coincidence site lattice (CSL) boundaries for rapid and stable grain growth. We expect these *in situ*

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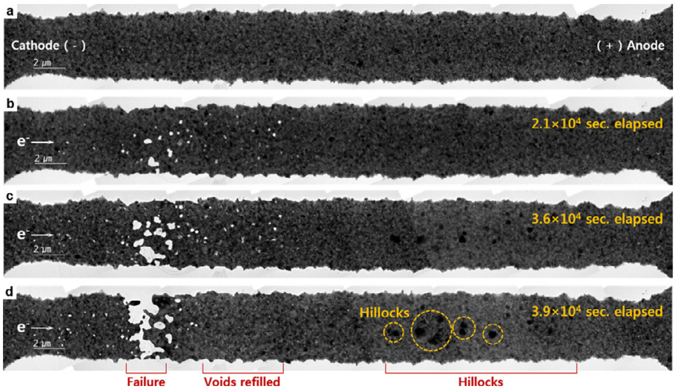

Fig. 1. Electromigration (EM) steps in a current-applied copper (Cu) line observed by *in situ* transmission electron microscopy (TEM). Bright-field images of the Cu interconnect were obtained (a) for the pristine state, and at (b) 350, (c) 600, and (d) 710 min after the current bias was fixed; the current density was 6×10^6 A/cm^2. The current pulse was held for 0.02 s and released for 10 s. Voids formed and widened near the cathode (b, c), eventually leading to interconnect failure (d). Dark spots near the anode side of the interconnect, marked with circles, indicate that hillocks formed at the same time.

experimental results to provide insight into the EM mechanism to advance the design of reliable interconnects by determining optimum fabrication parameter values.

2. Materials and methods

2.1. TEM sample preparation

A SiN$_x$ membrane (thickness: 50 nm) was deposited onto the SiO$_2$/Si substrate by low-pressure chemical vapor deposition. KOH-preferred etching of the Si substrate was conducted to make a self-supporting SiN$_x$ membrane, and a Cu film (thickness: 90 nm) was deposited via ultrahigh vacuum direct current (DC) magnetron sputtering. A line (width: 4–4.5 µm) was then added via photolithography and wet etching. A schematic diagram of the sample preparation procedure is shown in Figure S1(a).

2.2. In situ I–V measurement and TEM investigation

The patterned Cu line sample was loaded into a custom-built TEM stage with electrical feedthroughs and contacts to the Cu thin films. Electric currents were applied to the Cu lines in the TEM using an Agilent (B2912A) source meter. Pulses were applied with a duty cycle of 2×10^{-3} (on for 0.02 s/off for 10 s) and a current density of approximately 6×10^6 A/cm^2 to minimize thermal heating. To limit the electron beam effect during the migration process, the electron beam valve was closed except for the short period before observation of the first void. ASTAR mapping was then initiated. Pictures of the TEM holder and the prepared sample are shown in Figure S1(b). *In situ* TEM investigations were performed with a 200-kV field-emission TEM instrument (JEOL 2010F and 2100F). EM-induced microstructural changes of the Cu line in TEM were recorded using a slow-scan charge-coupled device (CCD) camera at one frame per second (fps) until 4×10^4 s. An ASTAR (NanoMEGAS) device and software were used to obtain crystallographic orientation data with the TEM instrument (JEOL 2100F).

3. Results and discussion

Fig. 1 shows the steps of interconnect failure due to EM processes observed using *in situ* TEM with external biasing. A pristine interconnect composed of polycrystalline 100-nm Cu grains was used (Fig. 1(a)); the fabrication procedure of the polycrystalline Cu line is detailed in the Materials and Methods section. When 6×10^6 A/cm^2 current density pulses were applied for approximately 6 h, voids nucleated on the cathode side of the interconnect (Fig. 1(b)). As we extended the biasing time, more voids formed (Fig. 1(c)), and some voids that had formed earlier were expanded. In addition, the number of hillocks formed near the anode, which appear as dark spots in Fig. 1, also increased over time. The formation of voids and hillocks is consistent with the known effects of EM [11–12]. The straight-line structure of the specimen removes the possibility of thermomigration [13], the mass-transport of elements driven by a thermal gradient, because no significant temperature difference would arise from the one-directional current flow in a straight line [14–15]. The local thermal gradient due to electron-beam heating may be a concern; however, the patterns of void and the hillock development were consistent throughout the repeat experiments performed without the electron-beam. In addition, we applied pulse currents of 0.02 s and 10 s for electrons to reach “flow” and “stop” states, respectively, to minimize thermal loading and prevent excessive temperature increases above the device-operating temperature, but maintain a high current density to accelerate the EM.

As voids widened and became connected, the resultant reduced cross-sectional area led to increased current density and, thus, an increase in the void expansion rate (Fig. 1(c)). Finally, the failure mode developed rapidly (Fig. 1(d)). Some of the small voids formed in the earlier stage filled up and disappeared as hillocks grew. To understand the EM mechanisms linked with microstructure, we recorded video of some of the voids for several hours while a pulsed current was applied. Before taking the series of snapshots, we obtained crystal orientation maps to correlate changes in grain orientation. Given that interconnect failure generally occurs with the aid of Cu self-diffusion on voids [1,3], the void expansion process at surfaces and grain boundaries, *i.e.*, paths where diffusion occurs most rapidly, were recorded (Fig. 1(d)).

A previous report suggested that the EM of Cu lines primarily occurs along the favorable orientation under surface diffusion at the device-operating temperature, with Cu atoms preferentially migrating on $\{111\}$ along $\langle 110 \rangle$ at the interface of voids and grains (notably the densest plane and direction of the Cu crystal, respectively)[8]. By observing at the atomic level, the same research group further elucidated in another report that a void first forms at high-angle grain boundaries by grain-boundary diffusion, and then expands via surface diffusion along particular orientations of free surfaces generated between the void and grains [9]. Our investigation, at scales from tens of nanometers to tens of micrometers, provides a link between atomic- and micron-level-scale observations.

Fig. 2(a)–(d) shows snapshots captured from time-compressed video (Supporting Information, Video S1). Red dashed lines indicate the boundary of the void shown in the prior image. Fig. 2(a)–(c) shows TEM bright-field images at 180, 195, and 340 min at a pulse current density of 6×10^6 A/cm^2, and Fig. 2(d) after an additional 8 min with the current density increased to 7×10^6 A/cm^2. Fig. 2(e) shows the color-coded crystallographic orientation map of the region at the early stage of void formation, acquired by ASTAR. The crystallographic orientation map was combined with a virtual bright-field image (contrast inverted), reconstructed from transmitted beam intensities while scanning nanobeam electron diffraction patterns [10]. Color codes are expressed using the standard stereographic triangle (Fig. 2e, inset). Dashed white lines indicate the corresponding void boundaries in the bright-field images (Fig. 2(a)–(d)). To trace preferential diffusion paths, the dominant volume reduction direction was measured from time-compressed video and marked with arrows (Fig. 2(e)). White and black arrows indicate the direction of Cu transport along grain boundaries and through the grain, respectively. The Cu transport direction was