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Engineering the grain boundary network of thin films via ion-irradiation: Towards improved electromigration resistance

Huan Ma [a], Fabio La Mattina [b], Ivan Shorubalko [b], Ralph Spolenak [a], Matteo Seita [c, *]

[a] Laboratory for Nanometallurgy, Department of Materials, ETH Zurich, Vladimir-Prelog-Weg 5, 8093 Zurich, Switzerland
[b] Laboratory for Reliability Science and Technology, Empa, 8600 Dübendorf, Switzerland
[c] Department of Materials Science and Engineering, Massachusetts Institute of Technology, MA 02139, USA

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ABSTRACT

Controlling the grain boundary network of small-scale materials—such as coatings and thin films—is an ambitious goal that would ultimately enable the production of components with tailored properties and improved reliability. In this work we present a new technique—which we term ion-induced grain boundary engineering (iGBE)—to engineer the character distribution and connectivity of grain boundaries in gold films *in situ*, as they are being deposited. iGBE consists of a repeated sequence of ion-induced material removal and material deposition which results in the selection and growth of crystal grains in twinned relationship. This phenomenon yields a substantial increase in the density and connectivity of Σ3 twin boundaries, which have renowned beneficial effects on the material's resistance to intergranular degradation. We confirm the improved reliability of iGBE microstructures by assessing their resistance to electromigration—one of the most common causes of failure in integrated circuit interconnects. We find that, through iGBE, interconnect lifetime increases by three orders of magnitude at standard operating conditions. Since iGBE is material-insensitive and compatible with standard microfabrication technology, we expect it to have significant impact on microelectronics industry.

1. Introduction

Most engineering materials are polycrystalline: they consist of an aggregate of differently oriented crystal grains joined along grain boundaries (GBs) [1]. Although they typically comprise a small fraction of the material's volume, GBs have tremendous impact on its properties, including strength and ductility [2–4], diffusion [5–7], resistance to environment-assisted failure [8,9] and radiation tolerance [10,11]. Indeed, by engineering the distribution and connectivity of GBs—namely the GB network—such properties can be improved by many orders of magnitude [12–15]. In metals and metal alloys, the GB distribution can be tailored by processing raw materials through specific thermo-mechanical treatments—a sequence of cold work and anneal—which take the name of *grain boundary engineering* (GBE) [16–20]. GBE yields an increased proportion of "special" GBs in the microstructure—primarily Σ3 boundaries—which have the effect of improving the material's performance and its resistance to intergranular degradation [21].

While GBE is largely employed for the fabrication of high-performance bulk materials, its use at the small-scale—such as on coatings or thin films—is still limited, due to the challenge of selectively deform and anneal small volumes of material [22]. Here we present a new method to engineer the GB network of as-deposited films and thus improve their performance. Our method consists of a combination of ion-irradiation and material deposition processes to selectively control the crystal grain orientation distribution, *in situ*, during sputter-deposition. We demonstrate the method—which we refer to as *ion-induced grain boundary engineering* (iGBE)—using gold (Au) films and we prove the beneficial effect of iGBE by assessing the material's resistance to electromigration (EM)—a GB-controlled degradation phenomenon which causes failure of interconnects in integrated circuits [23].

2. Experimental

Au deposition is performed by DC magnetron sputtering (PVD Products, Inc.) at room temperature on (100) silicon wafers coated with 50 nm of SiO_2 and 50 nm of Si_3N_4. To improve the adhesion of the film to the wafer, a 10 nm thick Ti layer is deposited prior to Au.

* Corresponding author.
E-mail address: matteos@mit.edu (M. Seita).

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The base pressure in the sputter chamber is better than 10^{-6} mbar. During deposition, the pressure is increased to 10^{-3} mbar by flowing pure argon (Ar) in the chamber and the silicon substrate is kept in rotation (with a speed of 12 rpm) in order to obtain a homogeneous film thickness. Ion-irradiation is carried out by means of a gridded ion-source (KRI KDC 40) with collimated and neutralized Ar^+ beam. The ion-source is installed directly in the sputter tool at a fixed angle of 35° with respect to the film surface normal. This configuration allows for *in situ* ion-irradiation of the deposited films. The ion-energy, ion-current, and irradiation time are kept constant throughout the experiments presented here at 1.2 keV, 5 mA, and 8 min, respectively. Upon irradiation, the base pressure in the chamber is increased to ~4×10^{-4} mbar and the substrate rotation is turned off.

The film microstructure is characterized by means of a high-resolution scanning electron microscope (SEM, Magellan 400 FEI) using both secondary electron (SE) and backscattered electron (BSE) detectors. Grain orientation information is assessed via electron backscatter diffraction (EBSD) using a FEI Quanta 200 FEG SEM. Only EBSD data with confidence index (CI) larger than 0.1 is considered. Quantities such as crystallographic texture spread, average grain size, and area fraction of specific grain orientations are measured from EBSD data using the OIM Analysis Software (EDAX, Inc). The GB character distribution and the triple junction (TJ) distribution are measured from cleaned EBSD data using MTex 3.4.1 [24]—a free software for the MATLAB environment. The cleanup procedure used is similar to others found in the literature [25–27] and consists of grain CI standardization, neighbor orientation correlation with level 1, and a single iteration of grain dilation using grain tolerance angle of 5° and minimum grain size of 2 pixels. GBs are classified according to the coincidence site lattice model [28], using Brandon's criterion [29,30]. The film grain structure is characterized along the cross-section using high-resolution SEM, transmission electron microscopy (TEM, FEI Tecnai F30 and FEI Talos F200X). The TEM analysis is carried out in both bright field (BF) and high-angle annular dark field (HAADF) modes. Cross-sections and TEM lamellae are prepared using a focused ion beam (FIB) microscope (NVision 40, Carl Zeiss).

EM tests are carried out using a four-point probe method [31] on arrays of Au lines, patterned from 500 nm thick films via lithography. The lines are 10 μm wide and 700 μm long. All lines are coated with a 200 nm SiN_x layer by plasma enhanced chemical vapor deposition (Plasmalab 80 Plus, Oxford Instruments) at 300 °C. EM tests are performed in a shielded wafer prober at controlled temperature, ranging from 125 to 200 °C. The current density used for all tests is 1.3×10^7 A/cm^2. All lines are tested until open circuit failure.

3. Microstructure control via iGBE

Fig. 1 illustrates the elements of iGBE via schematics as well as EBSD maps taken from representative samples. First, a 50 nm thick Au film—hereafter referred to as the *reference*—is sputter-deposited onto a substrate. The reference microstructure consists of columnar grains with an average size of ~60 nm and strong (111) fiber texture normal to the film surface. The fiber texture—a common feature in the microstructure of thin FCC films [32]— is clearly visible from the {111} pole figure in Fig. 1a, where the central peak and the continuous ring at ~70° indicate that all grains have the same surface normal, but random in-plane orientation, respectively.

After deposition, the reference film is irradiated at a fixed angle of 35° with respect to the surface normal (see schematics in Fig. 1b). As the energetic ions strike the film surface, two different phenomena take place in the crystal grains, depending on their orientation: *ion-milling* and *ion-channeling*. Grains that are randomly-oriented with respect to the ion-beam direction undergo ion-milling and thus are progressively removed from the sample surface [33]. Conversely, those oriented such that their open-most crystal-axis is parallel to the ion-beam undergo ion-channeling and thus survive ion-irradiation [34]. We refer to the combination of both phenomena as *selective grain milling* (SGM). The EBSD measurement of the reference after SGM is reported in Fig. 1b. Notably, the {111} pole figure now exhibits three symmetric peaks at ~70° from the normal, which suggests that all grains left on the surface have similar crystallographic orientation [35,36].

In FCC materials, such as Au, the easiest channeling-axes coincide with the ⟨110⟩ crystallographic directions [37]. Owing to the pre-existing (111) out-of-plane texture in the reference film, all grains in the microstructure exhibit three possible ⟨110⟩ channeling axes at an angle of 35° from the surface normal—exactly the angle at which the ion-beam is set. However, since the reference film is held steady during SGM, only one specific subset of grains is found in channeling-orientation and thus survives SGM. These are the grains appearing in the EBSD map in Fig. 1b. The orientation spread of the surviving grains is ~20°, as measured by EBSD. This value is in good agreement with the maximum angular deviation from the nominal channeling-axis that is allowed for the ion-energy, ion-species, and target material used in this study. In fact, following the model proposed by Lindhard [34], we compute a maximum deviation of:

$$\psi_c = \left(\frac{3a^2 Z_a Z_b e^2}{E_i d^3}\right)^{1/4} = 21^\circ \quad (1)$$

where Z_a and Z_b are the atomic numbers of ions and target atoms, respectively, $e^2 = 14.4$ eV Å (the square of the elementary charge), E_i the ion energy (1.2 keV), d the distance between two atoms along the ⟨110⟩ channel wall in Au (2.9 Å), and $a = a_B 0.8853(Z_a^{2/3} + Z_b^{2/3})^{-1/2}$, where a_B is the Bohr radius. The agreement between the measured orientation spread and ψ_c confirms that the grains remaining on the sample surface after SGM are in channeling-orientation and that all the other randomly-oriented grains were selectively removed. In our experiments, 8 min is the shortest irradiation time that yields complete removal of randomly-oriented grains. In fact, after 8 min the material's coverage on the sample surface is measured to be 25.3%, which is very close to the measured fraction of channeling-oriented grains in the reference film (25.7%, see Table 1), as well as to the predicted value in a fiber textured film (27.5%, see Ref. [35]).

In the next step of iGBE, a supplemental 50 nm Au film is sputter-deposited onto the irradiated reference without breaking the vacuum. The resulting microstructure—hereafter referred to as *1 × iGBE*—exhibits a higher fraction of channeling-oriented grains (~39.7%, see Table 1), as well as a large proportion of twinned grains (~26.4%, see Table 1). That can be seen from the pole figure in Fig. 1c, where the six-fold symmetry about the (111) fiber texture suggests the presence of two twinned orientations rotated by 60°. The increased fraction of channeling-oriented grains is due to the epitaxial growth of the pre-existing channeling-oriented grains—namely those which survived the first SGM [38]. At the same time, the favorable conditions for twin nucleation onto (111) crystal planes during physical vapor deposition of FCC materials leads to the preferential growth of grains in twinned orientation [39,40]. This means that upon the supplemental deposition of 50 nm of Au, the adatoms diffusing to the channeling-oriented grains can arrange themselves in two—equally favorable—crystallographic orientations: one which follows the parent orientation (namely the channeling-orientation), and one that is in