

Copper thin film of alternating textures

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It is common for thin films to have a predominant texture, but not alternating textures. In this letter, we report a copper film of alternating textures through self-organization. Using dc magnetron sputtering technique, we deposit copper films on a SiO₂/Si(111) substrate. A thin layer of copper of $\langle 111 \rangle$ texture is first developed, and another thin layer of $\langle 110 \rangle$ ensued. As deposition continues, a third layer of copper of $\langle 111 \rangle$ texture is formed on the top, leading to a sandwich copper thin film of alternating $\langle 111 \rangle$ and $\langle 110 \rangle$ textures. The film morphology is characterized with scanning electron microscopy and atomic force microscopy and the texture with x-ray diffraction. Based on anisotropic elastic analyses and molecular dynamics simulations, we propose a model of texture evolution during the formation of multilayers, attributing the texture evolution to the competition of surface and strain energies. © 2003 American Institute of Physics. [DOI: 10.1063/1.1583866]

Thin films usually are deposited with one dominant texture. Unless the deposition process is interrupted, the texture will not change through the entire film thickness. When layered structures are preferred, one generally resorts to multilayers consisting of different materials. Such multilayers are commonly used in electronic and magnetic applications.^{1,2} The advantages of these multilayers are well demonstrated by their widespread applications. It would be even better if the thickness of each layer can be changed after deposition: this may prove to be particularly advantageous for space applications. One possibility may be realized through the deposition of thin films of single-component materials, each layer having a unique texture. The thickness of each layer of given texture can be altered through texture competition.

Over the past years, we have been studying the texture competition by combining computer simulations³ and experiments.⁴ Recently, we have shown that $\langle 110 \rangle$ texture develops from a film/substrate of predominantly $\langle 111 \rangle$ texture during the deposition of copper thin films. In essence, the copper film deposited was a bilayer: one layer being $\langle 111 \rangle$ in texture and another $\langle 110 \rangle$. Simple elastic analyses indicate that it would be possible for the $\langle 111 \rangle$ texture to dominate again, if the deposition continues. In the following, we present the experimental realization of the return of the $\langle 111 \rangle$ texture, and thereby the formation of a sandwich copper film of alternating $\langle 111 \rangle$ and $\langle 110 \rangle$ textures; interpretation of the experimental results is facilitated by accompanying molecular dynamics simulations.

The copper film is deposited using dc magnetron sputtering technique. The experimental conditions are briefly summarized as the following. The sputtering power is 100 W, and the chamber is filled with 99.999% Ar, flowing into the chamber at a rate of 8.5 sccm. During the deposition, the base pressure is about 5.0×10^{-8} Torr and the working pressure is about 2.5×10^{-3} Torr. The target is a block of 99.995% copper, and is sputtering cleaned in Ar gas for 10 min before deposition. The substrate, which is 5 cm away from the target, is an *n*-type Si(111) wafer of resistivity 10–12 Ω cm. It can either be used as a clean Si substrate or oxidized before deposition, effectively making a SiO₂ substrate. In the latter case, the SiO₂ layers of about 0.1 μ m are the result of annealing at 300 °C in air for 1 h. In all cases, the substrate is cleaned ultrasonically in alcohol and then acetone.

Five samples of copper film, of various thicknesses, are prepared under the same deposition condition. They are deposited for 4, 7, 15, 25, and 35 min, respectively. Although the five thin-film samples are deposited separately, they should represent five snapshots in time, since they all are deposited under the same conditions. The scanning electron microscopy (SEM) images of the five snapshots are shown in Fig. 1 (left column). The corresponding atomic force microscopy (AFM) images are shown in Fig. 1 (right column). The x-ray diffraction (XRD) characterization is performed on each sample, and the relative intensities are shown in Fig. 2.

The SEM and AFM observations provide direct evidence of the morphological evolution of the copper films. During the early stage (first 4 min), the copper film is uniform. As the deposition proceeds (between 4 and 7 min), small hill-

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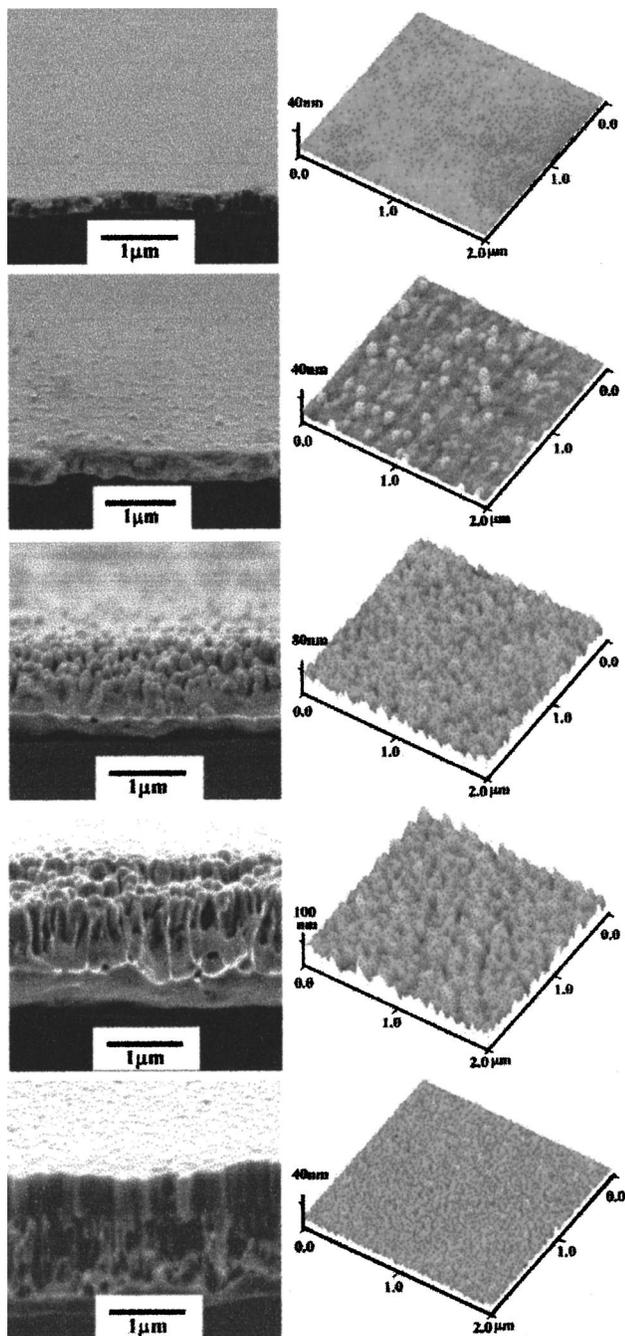


FIG. 1. Snapshot SEM (left) and AFM (right) images of copper films deposited on $\text{SiO}_2/\text{Si}(111)$ wafer at, from top to bottom, 4, 7, 15, 25, and 35 min.

ocks start to nucleate and grow from the uniform film. These hillocks grow into columns and thicken (within the first 15 min), and continue to develop into tall columns (up to 25 min). Finally, the columns collapse and the film returns to be uniform, and the surface is smooth again, as in the first snapshot.

The XRD characterization shows that the texture is evolving as the deposition proceeds. During the early stage, the uniform film is predominantly of $\langle 111 \rangle$ texture. As the hillocks develop, the $\langle 110 \rangle$ grains become more populous, indicating that the hillocks are of primarily $\langle 110 \rangle$ in vertical orientation. As the hillocks develop into columns and collapse, the relative fraction of $\langle 110 \rangle$ grains increases, as indicated by the increase of (220) relative intensity in Fig. 2. The

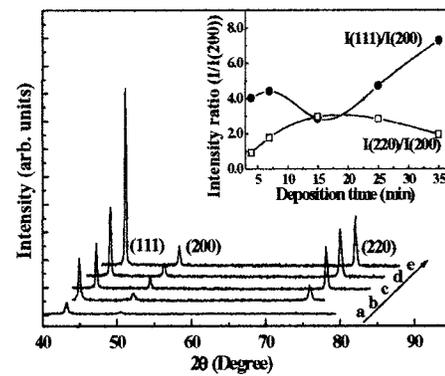


FIG. 2. XRD intensity of copper films deposited on $\text{SiO}_2/\text{Si}(111)$ wafer at (a) 4, (b) 7, (c) 15, (d) 25, and (e) 35 min. The inset shows the variation of relative intensities as a function of time.

$\langle 110 \rangle$ texture corresponds directly to the (220) intensity in XRD; the difference is only in terminology. At the final stage, when the film becomes uniform and the surface becomes smooth, the fraction of $\langle 111 \rangle$ texture regains its dominance. The relative intensities of $\langle 110 \rangle$ and $\langle 111 \rangle$ with respect to that of $\langle 100 \rangle$ are plotted as a function of time in the inset, to demonstrate more clearly the transition of texture with time. The change of XRD intensity indicates that the uniform film is of primarily $\langle 111 \rangle$ and the columns/hillocks are of primarily $\langle 110 \rangle$ textures. In other words, the final copper film shown in Fig. 1 (bottom) is a sandwich; the top and bottom layers are of $\langle 111 \rangle$ texture and the middle layer is of $\langle 110 \rangle$ texture.

As discussed in Ref. 4, the effective lattice constant of the initial $\langle 111 \rangle$ textured copper film is 0.2097 nm (about 0.5% strain), corresponding to stress of several hundreds of megapascals. This is beyond the yield stress of single-crystal bulk copper, and the difference in yielding⁵ could explain the $\langle 110 \rangle$ dominance over $\langle 111 \rangle$. However, the large strain/stress is observed in the thinnest film (top of Fig. 1). This strain is the net strain after dislocation activation, even if it did happen. Therefore, the anisotropy of elastic energy would play an important role; this is more so since dislocation activation (nucleation and propagation) can be more constrained in thin films than in the bulk.⁶ Our molecular dynamics simulations (not presented here) do show that elastic energy anisotropy can lead to $\langle 110 \rangle$ dominance over $\langle 111 \rangle$, under the strain of $\sim 0.5\%$ tension or compression.

Based on the elastic anisotropy, we propose a model of texture evolution during the multilayer formation, schematically shown in Fig. 3. During the early stage, the surface (and interface) energy dominates over elastic energy, and therefore the $\langle 111 \rangle$ texture develops to minimize the total energy. As the strain builds up, elastic energy starts to dominate over the surface energy, and therefore $\langle 111 \rangle$ grains lose out to the $\langle 110 \rangle$ grains. This transition is made more favorable by the fact that newly formed $\langle 110 \rangle$ grains are less likely to be strained (or so much strained). Therefore, $\langle 110 \rangle$ grains nucleate near $\langle 111 \rangle$ grain boundaries, as shown in Fig. 3 (second image from top). The geometrical shadowing effects in sputtering deposition give these newly formed $\langle 110 \rangle$ columns another niche; as a result, these columns grow even more. Finally, the columns collapse and form grain boundaries. At this point, strains may have developed in the $\langle 110 \rangle$

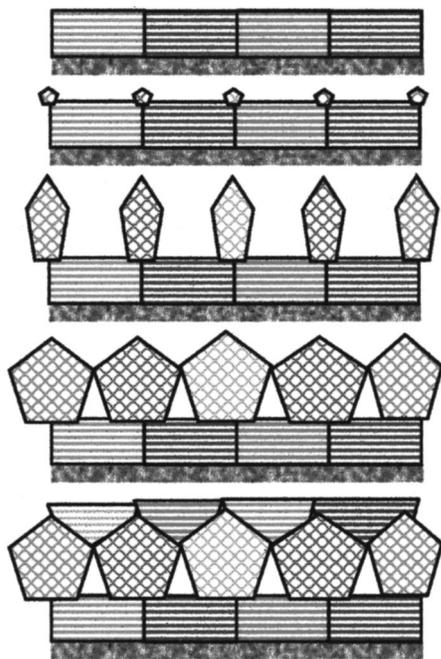


FIG. 3. Proposed model of texture evolution during the formation of copper multilayers, corresponding to the five snapshots in Fig. 1. The hatched lines indicate close-packed directions, and different gray scales indicate different grains. The bottom strip represents an amorphous substrate.

grains, due to the larger grain size and close proximity of other grains. More importantly, if any new $\langle 111 \rangle$ grains nucleate, they will be strain free (or at least less strained). The grain boundaries, as shown in Fig. 3 (fourth image from top), provide the necessary nucleation site of new $\langle 111 \rangle$ grains. Due to the absence of strain and smaller surface energy of the newly formed $\langle 111 \rangle$ grains, they grow at the expense of collapsed $\langle 110 \rangle$ grains, and form a new layer of uniform $\langle 111 \rangle$ grains. Before closing, we point out that this

model is meant to help interpret the experimental observation; we are not yet able to exclude other possible mechanisms at this point.

In summary, we have deposited a copper film of alternating $\langle 111 \rangle$ and $\langle 110 \rangle$ textures. The texture alternation during continuous deposition is a self-organization process. The mechanisms of such self-organization are not entirely understood. However, our proposed model, based on competition of strain and surface energies, does offer a reasonable explanation of the experimentally observed sandwich structure. If the proposed mechanism is valid, we anticipate that the alternation of $\langle 111 \rangle$ and $\langle 110 \rangle$ will continue beyond three layers, leading to multilayers of single-component materials. This possibility is being explored for copper and other thin films.

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