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Rapid annealing of metal thin films by micron chevron-shaped laser beam scanning

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Abstract

Micron chevron-shaped laser beam scanning was used for annealing of Al and Au thin films. Rapid grain growth was possible in solid state, but single-grain crystal growth via melting only succeeded for thermally deposited Al films, which had a sufficiently low reflectivity. Further optimization of the process to grow single-grain crystal on the sputtered Al films and the Au films was proposed and discussed.

1. Introduction

Metal thin films, in particular, Al and Au have found many applications in electronics and plasmonics. The metal thin films deposited by different methods are often polycrystalline with the grain size in the order of tens of nanometers.

The nano-grained structure with a high density of grain boundaries causes conductivity deterioration from significant contribution of grain boundary scattering. The grain boundaries also impose intricate resistance to focused ion beam milling, which makes incompatibility with fabrication of complex nano-circuits on the thin films.

Annealing is a common practice to grow the crystal grains for reduction of grain boundary density. However, post-deposition annealing by conventional heating is usually ineffective, because the grain growth in thin films stagnates when the grain size becomes several folds of the film thickness.

In this study, we used a laser scanning system with a micron chevron-shaped beam [1] to grow grain selectively on the Al and Au thin films, which were prepared by using either thermal deposition or sputtering deposition with variation of the films thickness. The relationship between the films parameters and the crystallization processes is presented and discussed.

2. Experimental procedures

Al thin films were deposited on quartz substrates by thermal and sputtering evaporation techniques. The thickness of the thermally deposited Al film was 60 ± 20 nm. The thickness of the sputtered Al films was varied from 15 to 300 nm, as an attempt to control the films reflectivity.

The Au films were also deposited on quartz substrates by thermal and sputtering depositions without adhesion layers, and with adhesion layers of either 5 nm MoO_3 or 5 nm Ti. A capping layer of 200 nm SiO_2 was also applied to a 50nm Au film to reduce the risk of ablation.

A laser scanning system with a micron chevron-shaped beam was used [1]. The laser source was a 1.2 W ultraviolet multi-mode laser diode of 405 nm wavelength. The apex angle of the chevron-shaped laser beam was 60^o and the beam width was approximately 10 µm.

The substrate with metal film was attached to the sample stage, which was placed at the right angle to the incident laser beam. The beam scanned the sample surface as the sample stage moved in its horizontal direction at a given speed from 1 to 40 mm/s.

After laser scanning, local crystal orientations of the annealed regions were measured by using electron backscatter diffraction (EBSD) technique. The EBSD measurements were carried out on JEOL 7001FA scanning electron microscope (FE-SEM) operating at 15 kV.

3. Results and discussions

The results are presented and discussed separately for the Al films and the Au films.

3.1 Annealing of Al thin films

For the thermally deposited Al films, ultralong single-grain crystals were obtained by laser annealing as shown in Fig.1. The width of the single grain crystal is comparable with the size of the chevron-shaped laser beam and the length is only limited by the laser scan distance.

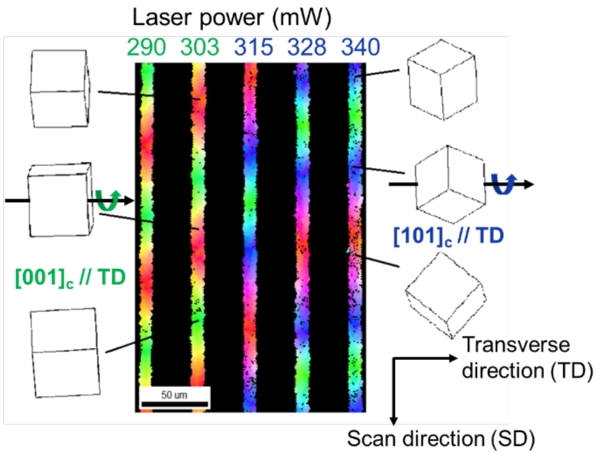

Fig. 1 EBSD orientation map showing the ultralong single-grain crystals obtained on the thermally deposited Al film. A positive pitch rotation of crystal orientation is observed for all the grains. The rotation axis changes from [001]//TD to [101]//TD as the laser power is increased.

All the single-grain crystals have a positive pitch rotation with a rotation rate of approximately 0.035 µm^{-1}. At laser power ≤303 mW the rotation axis is [001]//TD, while at the power ≥315 mW the rotation axis is [101]//TD. The relationship between the crystal quality and the laser scanning parameters were reported in detail elsewhere [2].

For the sputtered Al films of all thicknesses, the long single-grain crystals could not be obtained. A stable melting pool could not be maintained for single grain growth during laser annealing. At low laser power, the film did not melt, and only solid-state recrystallization occurred. At high laser power, the film melted but evaporated immediately due to high temperature of the melt.

Measurement of the reflective spectra of the Al films revealed that the thermally deposited Al films have abnormally low reflectivity (68%) at the laser wavelength of 405 nm. At the same wavelength, the sputtered Al films have high reflectivity (82-95%), which was loosely dependent on the films thickness or the film surface roughness.

Probably reflectivity is the key factor for single-grain crystallization of the Al films by laser annealing. It requires reducing reflectivity by optimization of the sputtering process or by optimization of the laser wavelength.

3.2 Annealing of Au thin films

For Au thin films, the reflectivity was low (47% at 405 nm), so that a sufficiently large amount of energy was absorbed by the films. However, only solid-state crystallization was obtained with the mean grain size increased ten folds and formation of a strong (111) crystal texture (Fig. 2). The width of the thermal affected zone is serval times of the laser beam size (Fig. 2b), but a stable melting pool could not be maintained for all types of Au films.

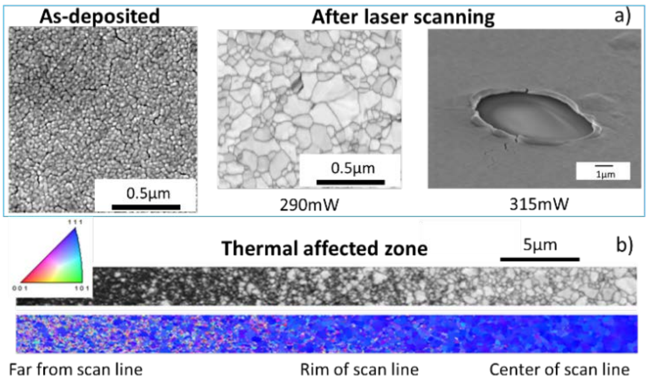

Fig. 2 Solid-state recrystallization of the Au films after laser annealing. a) mean crystal grain size increased ten folds at laser power 290 mW, but ablation occurred at laser power 315 mW. b) The thermal affected zone with extensive crystal growth at the center of the laser scan line.

We supposed that the Au liquid phase evaporates at a high rate judging from a high value of vapor pressure 2.28×10^{-3} Pa at its melting point 1337K. In order to stabilize the Au liquid pool, we have tried to deposit 200 nm SiO_2 capping layer on top of a 50 nm Au film to protect the melt from evaporation.

The capped Au film was annealed with variation of the laser scanning speed and laser power. Fig. 3 shows the EBSD image quality map and EBSD crystal orientation map of the capped film after 190 mW laser scan at 20 mm/s speed. Obviously, a continuous melting pool was formed at the center of the scan line. The width of the melting pool is half of the beam's size. However, upon solidification lateral grain growth took place resulting on a structure of elongated grains at an inclined angle to the scan direction.

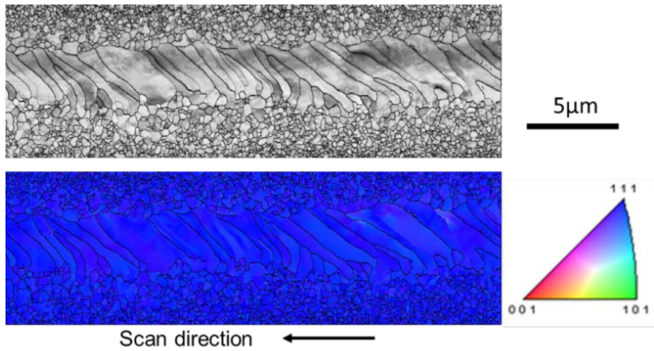

Fig. 3 EBSD image quality map and crystal orientation map of the 50 nm Au film capped with 200 nm SiO_2 after laser annealing. A melting pool was formed at the center, where lateral coarse grains crystallized with a strong (111) crystal texture.

Application of 200 nm SiO_2 capping layer is effective for suppression of evaporation, but the sandwich film structure altered the reflectance of the Au film so that the Au film was melted at a low laser power of 190 mW. This led to a narrow melting pool and promoted lateral grain growth. Further optimization of the capping layer materials or the layer's thickness is required to grow single-grain crystal on Au thin films.

4. Conclusions

Micron chevron-shaped laser beam scanning is effective for selective and rapid annealing of Al and Au thin films, which could result in ten folds of grain size increase and strong (111) crystal texture via solid-state recrystallization.

For thermally evaporated Al film with sufficiently low reflectivity, ultralong single-grain crystal can be obtained, but for sputtered Al thin film, further optimization of the process is required for single-grain crystal growth.

For Au thin films, a stable melting pool was obtained with the use of 200 nm SiO_2 capping layer to suppress evaporation, but the single-grain growth still is a challenge.

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References

[1] W. Yeh *et al.*, Single-grain growth in Si film by chevron-shaped cw laser beam scanning, Applied Phys. Exp. 9 (2016) 022503 (1-4).
[2] A.H. Pham *et al.*, Selective growth of single-grain crystal in Al thin film by micron chevron-shaped laser beam scanning, Thin Solid Films, 672 (2019), 100-103.