様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

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研究課題名（和文）Selective growth of ultra-long grain in metal thin film for microelectronic application

研究課題名（英文）Selective growth of ultra-long grain in metal thin film for microelectronic application

研究代表者

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研究成果の概要（和文）：レーザービーム走査を用いて、熱蒸着したAl薄膜とスパッタ蒸着したAu薄膜上に単結晶結晶縞を成長させることに成功した。 レーザービーム走査により、Au薄膜上に単粒結晶を成長させるために必要な条件を確立した。 金属薄膜上での単結晶結晶成長の場合、過度のアブレーションなしに完全な溶融プールを維持するようにレーザービームを制御する必要がある。 単粒結晶成長のメカニズムとその結晶方位選択を解明した。

研究成果の学術的意義や社会的意義
We have grown single-grain crystal line on Al and Au thin films. The laser parameters have been determine to control the crystal orientation of the crystals. The mechanism of crystal orientation selection has been clarified. The results have applications in microelectronic and plasmonic devices.

研究成果の概要（英文）：In this research project, we have successfully grown single-grain crystal stripes on thermal-evaporated Al thin films, and sputter-deposited Au thin films by using laser beam scanning. We have established the conditions necessary for the growth of single-grain crystal on Au thin film by laser beam scanning. For single-grain crystal growth on metal thin films, the laser beam should be controlled to maintain a complete melt pool without excessive ablation. A SiO2 capping layer thicker than 400 nm is required. The mechanism of single-grain crystal growth and its crystal orientation selection was elucidated.

研究分野：Thin film

キーワード：Thin film　Single Crystal　Laser annealing　Metal film　Crystal orientation　Crystal growth　Microstructure control　Characterization

様　式　C－１９、F－１９－１、Z－１９（共通）

１．研究開始当初の背景

Metal lines of Al, Cu or Au are important components in microelectronics. They are used as interconnect lines in integrated circuits, as electrodes in organic light-emitting diode or surface acoustic wave devices. One big problem with the thin metal lines is that their electrical conductivity and their durability depend inversely on the density of grain boundary. It is because of the dominance of grain boundary scattering and grain boundary diffusion in thin lines under electric current, which leads to a famous failure mechanism called electromigration.

A boundary-free metal line made of a single-grain is probably ideal for electronic application. However, as the metal lines are often fabricated from metal thin films which have ultra-fine grain size of few hundred nanometers, the density of grain boundary in the metal lines is very high. Currently post-deposition annealing can only increase the grain size to few micrometers, which is not sufficient to achieve single-grain lines. Laser annealing has been widely used for single-grain growth of semiconductors in amorphous thin film, but a similar approach to metal thin films has not been reported so far.

２．研究の目的

This proposal focuses on the problem of single-grain growth in metal thin film by laser annealing. The key scientific question is by what condition a single-grain line can be grown in metal thin film during laser annealing. The three specific aims of this proposal are therefore:

1) The effect of laser annealing on grain growth of metal thin film

Hypothesis: As for semiconductor, the grain growth of metal thin film during laser annealing depends on the laser beam shape, beam energy, the film thickness and crystal structure of the metal.

Expected result: Establishment of parameters dominating the grain growth during laser annealing and relationship between the parameters and the annealed microstructure.

2) Mechanism of grain growth during laser annealing

Hypothesis: Recrystallization of the metal thin film may occur in solid or liquid states. The mechanism may be different with variation of the beam parameters and the microstructure of the film.

Expected result: Mechanisms of grain growth by laser annealing of thin films of Al and Au.

3) Controlling crystallographic orientation of the recrystallized grain

Hypothesis: Some specific crystal orientations are preferable for single-grain growth. The selection of one crystallographic orientation depends on surface energy, liquid-solid interface energy, temperature profile of the melt pool, which depends on the energy of the laser beam.

Expected result: Optimization of the laser annealing process in order to grow a single-grain of a desired crystallographic orientation.

３．研究の方法

Al thin films were deposited at $5.0\text{x}10^{-4}$ Pa vacuum on a quartz substrate (18 mm x 18 mm x 0.2 mm) by thermal evaporation method. The thickness of Al film was 60 ± 20 nm measured by atomic force microscopy. The 50 nm Au films on 15 mm x15 mm x0.18 mm borosilicate glass substrate, which were purchased from PHASIS (Switzerland), were deposited by radio-frequency (RF) magnetron sputtering method. A 5 nm Ti layer was used to increase the adhesion between the Au film and the glass substrate. A SiO_2 capping layer was deposited on top of the Au film by reactive pulse DC magnetron sputtering to suppress ablation of the Au during laser annealing. The thickness of the SiO_2 layers was 200, 300, 400, and 800 nm.

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 side of the chevron shape was approximately 10 μm.

The metal films were 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 10 to 40 mm/s. After laser scanning, the 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.

４．研究成果

Ultra-long, single-grain crystal of 9-μm width can be grown selectively in Al thin film by laser scan annealing. The crystal quality and the morphology of the crystal depend on the laser power, scan patterns, and the scan speed as shown in Fig. 1a. The crystal orientation continuously rotates in a positive pitch rotation. The rotation axes are either $[001]_c$ // TD or $[101]_c$ // TD depending on the laser power. The transition between the two rotation axes is at the laser power of 303-315 mW.

Because the laser beam is well focused during the scan path, an ultra-long single grain can be obtained with the length comparable with the scan distance. A wide-area of single-grain stripes can also be realized by a 2D scan as shown in Fig. 1b and 1c. The obtained results would have potential applications in microelectronic and plasmonic devices.

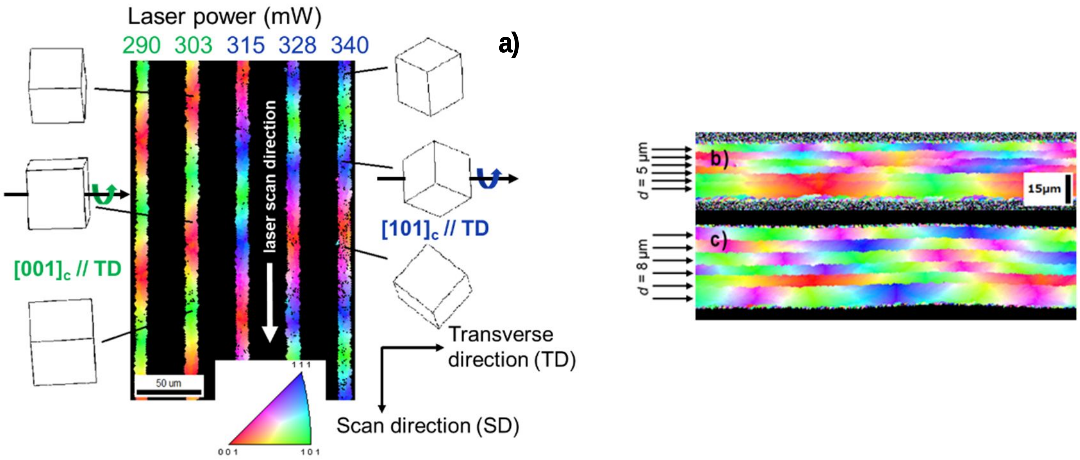

Fig. 1. (a) Crystal orientation maps of the single-grain crystal stripes obtained by scans with variation of laser power. The cubes showing the crystal rotation along the laser scan direction. (b) and (c) are crystal orientation maps obtained after wide-area annealing (2D scan) by repetition of six scans with an open interval *d* between the scan line centers. The arrows indicating the laser scan direction are placed at the center of every scan lines.

For the Au thin film, the single-grain crystal could be grown, when a sufficiently thick SiO_2 capping layer was deposited on the Au film to prevent ablation during laser scanning. The orientation of the obtained single-grain crystal in Au film varied with the thickness of SiO_2 capping layer, the laser scan speed, and the laser power.

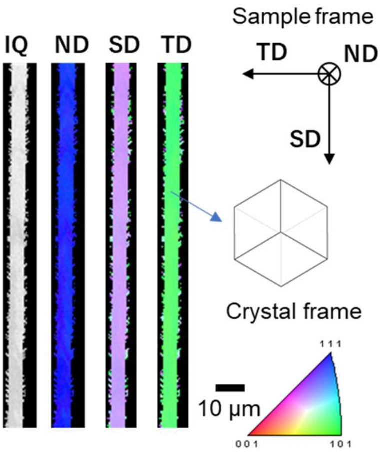

Fig. 2. EBSD image quality map (IQ) and crystal orientation maps plotted from the sample normal direction (ND), laser scan direction (SD), and transverse direction (TD) for a single-grain crystal obtained on Au thin film by laser beam scanning. The crystal orientation was [111]//ND, $[11\bar{2}]$//SD, and $[1\bar{1}0]$//TD.

Fig. 2 shows EBSD image quality map and typical crystal orientation maps of a single-grain crystal obtained on the Au thin film with 800 nm SiO_2 capping layer. The width of the single-grain stripes varied between 6-9 µm, depending upon the laser scanning parameters. The length of the stripes was comparable with the distance of laser scanning. All the single-grain crystal stripes have (111) plane orientation, which was selected to minimize their surface energy. 77% of the stripes have the in-plane [112] direction aligned with the laser scan direction (SD). The selection of [112] // SD can be explained if the anisotropy of surface tension was accounted for. In the (111) plane of the Au film, the surface tension is maximum in the <110> directions, which are favorable for the crystal growth. The stripe will grow preferentially with its two <110> directions close to the thermal gradients, which are symmetrically inclined with respected to the laser scan direction. The [112] direction is the sum of those two symmetrical <110> directions, hence it is aligned with the SD direction. The scenario is similar to the growth of degenerate seaweed dendrite observed in one-directional solidification of Al alloys.

５．主な発表論文等

〔雑誌論文〕　計2件（うち査読付論文　2件／うち国際共著　2件／うちオープンアクセス　0件）

１．著者名 Pham Anh Hoang、Fukunaga Naruki、Yeh Wenchang、Morito Shigekazu、Ohba Takuya	４．巻 60
２．論文標題 Rapid annealing of Au thin films by micron chevron-shaped laser beam scanning toward growth of single-grain crystal	５．発行年 2021年
３．雑誌名 Japanese Journal of Applied Physics	６．最初と最後の頁 SBBK06～SBBK06
掲載論文のDOI（デジタルオブジェクト識別子） 10.35848/1347-4065/abdf22	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

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掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.tsf.2019.01.023	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

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１．発表者名 A.H. Pham, N. Fukunaga, W. Yeh, S. Morito, T. Ohba
２．発表標題 Rapid annealing of metal thin films by micron chevron-shaped laser beam scanning
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〔図書〕　計0件

〔産業財産権〕

〔その他〕

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６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関