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Importance of MoS_2-Compound Sputtering even with Sulfur-Vapor Anneal for Chip-Size Fabrication

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Abstract

High quality MoS_2 film was synthesized by RF magnetron sputtering and sulfur-vapor annealing. The crystallinity of MoS_2 film was controlled by sputtering conditions. And we revealed a relationship of the crystallinity between as-sputtered and sulfur-annealed MoS_2 films. It is found that a crystallinity improvement after the sputtering is mandatory to achieve the excellent quality of MoS_2 film after sulfur-vapor anneal.

1. Introduction

Molybdenum disulfide (MoS_2) film is a promising material for human interface FET applications, because of its transparency, flexibility and high mobility (~ 380 cm^2/V-s) even in atomic thickness [1,2]. The MoS_2 film with continuous large area and high quality is essential for the high performance of MoS_2 device. For these preconditions, several research groups have synthesized the CVD-MoS_2 film using perylene-3,4,9,10 tetracarboxylic acid tetrapotassium salt (PTAS) to promote lateral growth, which influences an intrinsic MoS_2 properties [3]. On the other hand, a sputtered-MoS_2 film is superior to others in terms of uniformity, chip-size deposition and high thickness controllability. It has been reported that MoS_2 film was formed by using Mo- or MoS_2-target sputter and sulfurization [4-6]. It is easily speculated that the MoS_2 target sputter is better than Mo one, because MoS_2 one leads higher quality of final MoS_2 film through metastable state of just-after-sputtered MoS_2 film. Although sputtered-MoS_2 film has been investigated by controlling the sputter parameters [7,8], the high quality of MoS_2 film in metastable state is expected as high as possible to obtain the fine final MoS_2 film.

In this work, we investigate the further improvement of MoS_2 film quality with discussing on the film crystallinity before and after sulfurization.

2. Experiments

MoS_2 films were formed by RF magnetron sputtering with 4N-MoS_2 compound target on SiO_2/n-Si substrate, as shown in Fig. 1 (a). In order to prepare various MoS_2 films, the sputtering conditions were changed as RF power of 30 to 50 W, substrate temperature of 200 to 500ºC under Ar pressure of 0.55 Pa, Ar flow rate of 7 sccm and target-substrate distance of 150 mm. Figure 1 (b) shows a sulfur vapor annealing apparatus for sulfur compensation to MoS_2 film. The sulfur powder was placed at the zone1 heated at 250ºC for 40 min, and the samples were placed at zone2 heated at 700ºC for 40 min. The MoS_2 films were analyzed by the Raman spectroscopy for which the full width at half maximum (FWHM) values of A_{1g} and E_{2g}^{1} peaks were extracted.

3. Results and Discussion

Figure 2 shows A_{1g} and E_{2g}^{1} FWHM values of MoS_2 films varying RF power. The crystallinity improves with an increase in RF power up to 40 W, in which particle's energy enhances migration on substrate. On the other hand upper than 40 W, the degradation of crystallinity is seen, in which higher particle's energy and the large number of species cause sulfur defects and increment of nucleation density, respectively. This trend is also seen even after S-annealing.

Figure 3 shows FWHM values depending on substrate temperature. For as-sputtered MoS_2 film below 300ºC, MoS_2 crystallinity was improved along with an increase in the substrate temperature. However, the as-sputtered MoS_2 crystallinity deteriorated with higher than 300ºC. Figures 4 (a) and (b) show the X-ray photoelectron spectroscopy (XPS) of Mo *3d* for the as-sputtered MoS_2 films at 500 and 300ºC, respectively. Mo-Mo bonds are observed corresponding to sulfur defects due to an excessive sulfur desorption [7]. On the other hand at 300ºC, there is no peak of Mo-Mo bonds and Mo-O bonds, which is expected to be sulfurized by following SVA process.

In Fig. 3 with S-annealed data, FWHM values depending on temperature are almost the same as as-sputtered data. The excessive sulfur desorption might influence crystal recovery during sulfur annealing. From the X-ray diffraction (XRD) shown in Fig 5, (002) peak of MoS_2 film are found and the crystallites of MoS_2 is improved after sulfur annealing.

Figure 6 shows the correlation between FWHM values before and after sulfur annealing, based on the data in Figs. 2 and 3. All MoS_2 films except 400 and 500ºC are improved along with correlation coefficient of one. From this result, the improvement of as-sputtered film quality directly affects to the MoS_2 crystallinity after S-annealing, as shown in Fig. 7. Based on these discussions, a two-step process consisting of compound sputter using MoS_2 target and sulfur annealing is appropriated for the MoS_2 film improvement.

4. Conclusions

The crystallinity of the MoS_2 were controlled by sputtering conditions, and we demonstrated the mechanism of controlling the MoS_2 crystallinity in various sputtering conditions. The crystallinity of the sputtered MoS_2 film before S-annealing was retained after S-annealing. It was revealed an importance of improvement of compound sputtering even after sulfur compensation process. Further high performance of FET using thin MoS_2 film is expected to be applied for human interface FET applications.

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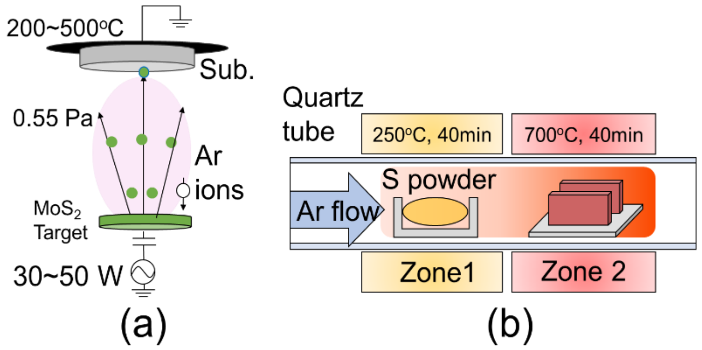


Fig. 1. Schematic diagrams of (a) RF sputtering system and (b) sulfur vapor annealing process tool.

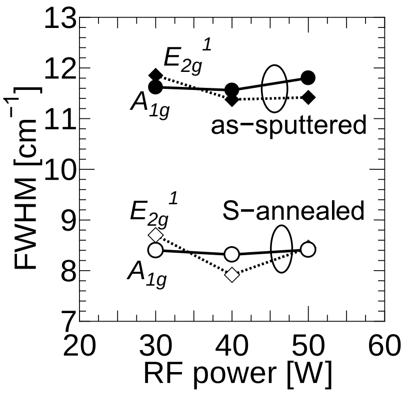


Fig. 2. FWHM values in $E_{2g}{}^{1}$ and A_{1g} modes from the Raman spectra for as-sputtered and S-annealed MoS_2 films depending on RF power, 0.55 Pa ,300°C.

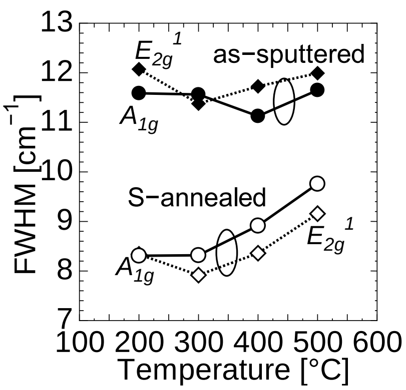
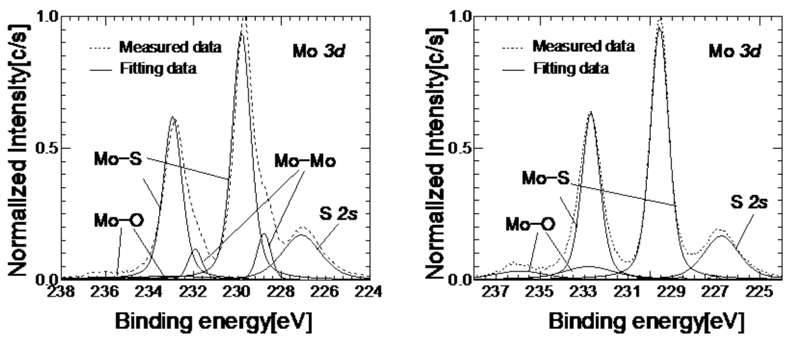


Fig. 3. FWHM values in $E_{2g}{}^{1}$ and A_{1g} of the Raman spectra for as-sputtered and S-annealed MoS_2 films depending on substrate temperature.



(a) (b)

Fig. 4. XPS spectra of molybdenum *3d* in as-sputtered MoS_2 film with the substrate temperature of (a) 500°C and (b) 300°C, RF power of 40 W and 0.55 Pa in Ar gas.

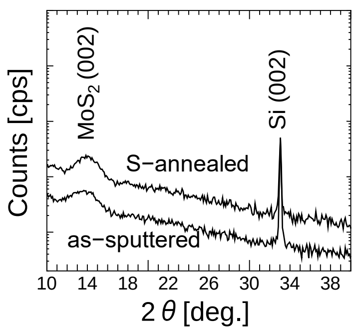


Fig. 5. XRD pattern of as-sputtered and S-annealed MoS_2 film. The films were sputtered with 40 W, 0.55 Pa at 300°C. Intensities have been normalized by silicon peak.

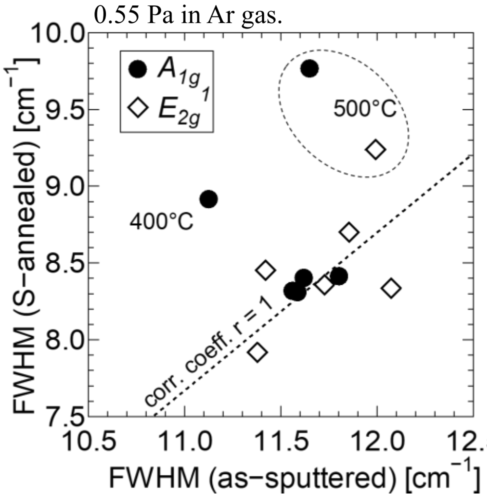


Fig. 6. Relationship of FWHM values in A_{1g} and $E_{2g}{}^{1}$ modes between as-sputtered and S-annealed MoS_2 films.

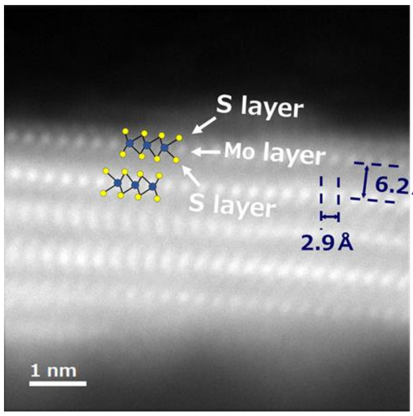


Fig. 7. Cross-sectional HAADF-STEM image of sputtered-MoS_2 film with 40 W at 300°C after sulfur annealing.