Centimeter-scale high-performance few-layer MoS₂ fabricated by RF magnetron sputtering and subsequent post-deposition annealing

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Abstract

High-quality few-layer MoS₂ films were fabricated by RT sputtering and subsequent post-deposition annealing in the same vacuum chamber. The fabricated films were characterized by XPS, Raman spectroscopy, Tauc plot analysis, and Hall effect measurements. As a result, it was confirmed that the films showed cm-scale uniformity, indirect bandgap of 1.1~1.2 eV, and superior electrical property of high Hall mobility than 36 $cm^2V^{-1}s^{-1}$ which is applicable enough for the future MoS₂ TFT LCDs with 8k resolution.

1. Introduction

Molybdenum disulfide (MoS₂), one of the two dimensional materials, is expected to be used in the future nanoscale devices owing to its novel physical, optical, and electrical properties. The properties are largely depended on thin film fabrication methods; especially, clean fabrication process is needed to realize normally-off MoS₂ devices [1, 2]. RF magnetron sputtering can produce cm-scale uniform MoS₂ thin film and is suitable to fabricate low-impurity high-performance thin-film transistor (TFT) channel. We previously reported that the few-layer MoS₂ fabricated by the high-temperature (HT) sputtering and subsequent sulfurization annealing showed the high performance applicable for 8k liquid crystal display (LCD) performance with normally-off operation [3]. In this study, cm-scale few-layer MoS₂ films were fabricated by room-temperature (RT) sputtering and subsequent crystallization annealing in the same vacuum chamber, which is a simple and clean process compared to the other existing methods. We also investigated the physical, chemical, and electrical properties of the films.

2. Results and Discussion

Fig. 1 shows an overview picture of few-layer MoS_2 fabricated on 1×1 cm c-plane sapphire substrate and process flow for the sample fabrication. Few-layer MoS_2 films were fabricated on 1×1 cm c-plane sapphire substrates by RF magnetron sputtering at RT and subsequent post-deposition annealing (PDA) in the same vacuum chamber at 400-600°C. The MoS_2 target is 99% pure and the base pressure is $< 2.0 \times 10^{-5}$ Pa. The thickness of the films were controlled by sputtering duration (27.8 s) as 10

layers (10L), which were confirmed by X-ray reflectivity measurement.

Fig. 2 shows Mo 3d XPS spectra of the as-deposited and the post-deposition annealed (600°C) 10L MoS₂ films. Mo-S chemical bonding peaks were observed in the both films; however, the peaks of as-deposited film were broadened due to the presence of native oxides and 1T-MoS₂ domains [4, 5]. After the annealing, it was considered that the films showed an ideal 2H-MoS₂ phase and were suppressed oxidation by the air exposure thanks to their high crystalline quality. S/Mo ratio of the as-deposited and the post deposition annealed 10L MoS₂ films were investigated by XPS multipoint measurements as shown in Fig. 3. We previously reported that the S/Mo ratios were decreased with increasing the substrate temperature for the HT sputtered MoS₂ films [6]. Contrastingly, the S/Mo ratio of the films after PDA showed constant value in all the measurement points, suggesting suppression of sulfur desorption. Fig. 4 shows Raman spectra of the post-deposition annealed (600°C) 10L MoS₂ film obtained by Raman multipoint measurements. The two Raman characteristic peaks, E¹_{2g} and A1g, were clearly observed and the peak positions, which were calibrated by the substrate peak positions labelled with an asterisk, remain identical in all the spectra, indicating that thickness and crystalline quality of the film were uniform over a cm-scale region. Then, we performed the bandgap estimation of the post-deposition annealed 10L MoS₂ films from Tauc plot analysis of the absorption coefficients [7], which were obtained by spectroscopic ellipsometry measurements, as shown in Fig. 5. The bandgap of the films increased with increasing PDA temperature and showed bulk MoS₂ value for the film annealed at 600°C. It was considered that the bandgap narrowing of the films annealed below 500°C were caused by residual 1T-MoS₂ phase in the films. Fig. 6 shows Hall mobility and carrier density of the post-deposition annealed and the post deposition sulfurized 10L MoS₂ films [3] obtained by Hall effect measurements. As mentioned above, 10L MoS₂ fabricated by HT sputtering and subsequent sulfurization annealing showed sufficiently high mobility and low carrier density [3]. For the PDA films, the Hall mobility was further improved. The Hall mobility value than 36 cm²V⁻¹s⁻¹ will contribute to realize future MoS₂ TFT LCD applications with 8k resolution.

3. Conclusions

High-quality 10L MoS₂ films were fabricated by RT sputtering and subsequent *in-situ* crystallization annealing. As a result, it was confirmed that the films showed cm-scale uniformity, indirect bandgap of $1.1 \sim 1.2$ eV, and superior electrical property, the high Hall mobility than 36 cm²V⁻¹s⁻¹ applicable for the future MoS₂ TFT LCD applications with 8k resolution.



Fig. 1 An overview picture of cm-scale few-layer MoS_2 fabricated on 1×1 cm c-plane sapphire substrate at 600°C and process flow for the sample fabrication.



Fig. 2 Mo 3d XPS spectra of the as-deposited and the post-deposition *in-situ* annealed (600° C) 10L MoS₂ films.



Fig. 3 S/Mo ratio of the as-deposited and the post deposition annealed 10L MoS_2 films (1 × 1 cm) obtained by XPS multipoint measurements (point-to-point distance: 2.5 mm).

References

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Fig. 4 Raman spectra of the post-deposition annealed (600°C) 10L MoS₂ film (1 \times 1 cm) obtained by Raman multipoint measurements (point-to-point distance: 2.5 mm).



Fig. 5 Bandgap estimation of the post-deposition annealed $10L MoS_2$ films from Tauc plot analysis.



Fig. 6 Hall mobility and carrier density of the post deposition annealed (PDA) and the post deposition sulfurized (PSA) 10L MoS_2 films [3].