ミスト CVD 法による 2D MoS_2, $WS_{1-x}Se_x$ 粒径拡大の検討

Synthesis of 2D MoS_2 and $WS_{1-x}Se_x$ thin films by Mist-CVD

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1.Introduction: We have studied the synthesis of 2D MoS_2 atomic layer thin films by mist CVD from ammonium tetrathiomolybdate $(NH_4)_2MoS_4$ using layer-by-layer (LbL) method. Previously, we presented (1) the number of stacking layers of MoS_2 flakes under the LbL mode was varied from atomic mono layer to 4− 5 layers by varying t_2, repetition cycles of precursor mist supply *N*, and solution precursors concentration. (2) The average size of atomic MoS_2 monolayer flakes deposited using LbL mode on *th*-SiO_2 and ATO dielectric layers increased to 20- 30 and 60-70 µm, respectively with relatively uniform size distribution. (3) Atomic MoS_2 monolayer-based FETs exhibited a mobility of 31- 40 (43- 55) cm^2/Vs with a V_{th} of -1.6 (-0.5 V), an on/off ratio of 3.0×10^4 (3.5×10^5), and SS of 0.80 (0.11) V/dec. for *th*-SiO_2 (ATO) gate insulator layers without mechanical exfoliation [1]. In this study, we attempt to further increase in the crystalline grain size of MoS_2 flake and extend the fabrication of WS_2 based ternary alloy, $WS_{1-x}Se_x$ thin layers by LbL method.

2.Experimental: $(NH_4)_2MoS_4$ and ammonium tetrathiotungstate $(NH_4)_2WS_4$ were used as a precursor of MoS_2 and WS_2 respectively and N-methyl-2-pyrrolidone (NMP) was solvent. MoS_2 and WS_2 films were synthesized by mist CVD with Ar gas containing H_2 (25%) at a T_f of 400- 600 °C on atmospheric pressure He plasma exposed *th*-SiO_2 and mist CVD grown $Al_{1-x}Ti_xO_y$ (ATO) substrates for 30 s. Subsequently, the sulfurization (selenization) of MoS_2 (WS_2) was executed for further improving the quality of the flakes at T_f of 600 °C for 20 min.

3.Results and Discussion: Fig.1a shows the camera, optical microscope and AFM images of MoS_2 precursor solution on ATO before and after exposing He plasma. The contact angle decreased from 29° to 21° by plasma exposure on *th*-SiO_2 and ATO substrates, suggesting that the wettability of deposition precursors was promoted. The optical microscope and AFM images of MoS_2 layers grown by LbL modes on ATO substrate before and after exposing He plasma are shown at the bottom. The average flake size of MoS_2 increased to ~70 to >100 µm on ATO substrate. Figure 1b shows the PL mapping of the corresponding MoS_2 thin layers on ATO with plasma exposure. The average size of atomic MoS_2 monolayer flakes increased from ~95 µm (continuous) to ~110 µm (LbL) with better uniformity than the continuous mode. This is revealed by the AFM thickness profile and the color contract of the PL mapping of LbL MoS_2 flake (Fig. 1b), which shows the formation of atomic mono/bilayer MoS_2 throughout the substrate surface. We also extend the synthesis of binary alloy $WS_{1-x}S_x$ and WSe_2 through the selenization of LbL grown WS_2.

[1] A. Kuddus et al. ACS Appl. Nano Mater. 2021 (to be published).

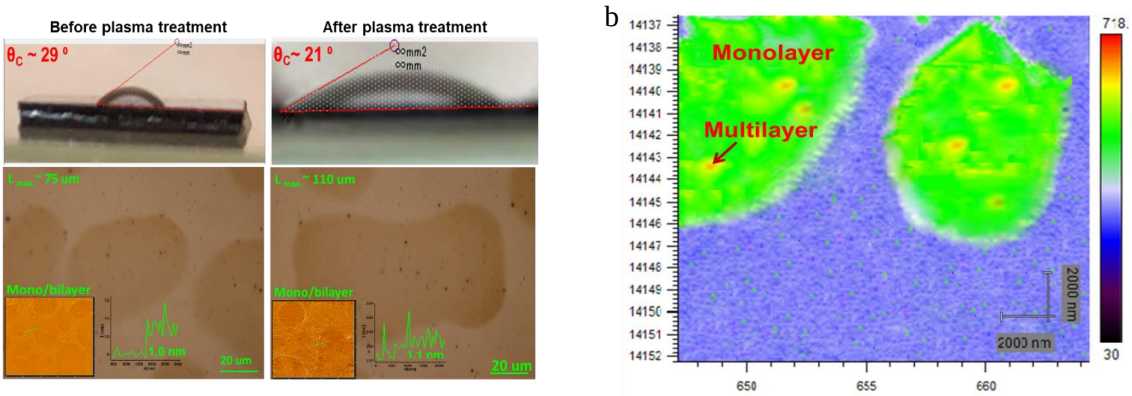

Fig.1 (a) Camera, optical microscope and AFM images of mist CVD MoS_2 with He plasma exposure. (b) 2D map of PL of atomic MoS_2 monolayer film on plasma exposed ATO.