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Large-area, continuous and high electrical performances of bilayer to few layers MoS₂ fabricated by RF sputtering via post-deposition annealing method

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We report a simple and mass-scalable approach for thin MoS₂ films via RF sputtering combined with the post-deposition annealing process. We have prepared as-sputtered film using a MoS₂ target in the sputtering system. The as-sputtered film was subjected to post-deposition annealing to improve crystalline quality at 700 °C in a sulfur and argon environment. The analysis confirmed the growth of continuous bilayer to few-layer MoS₂ film. The mobility value of ~29 cm²/Vs and current on/off ratio on the order of ~10⁴ were obtained for bilayer MoS₂. The mobility increased up to ~173–181 cm²/Vs, respectively, for few-layer MoS₂. The mobility of our bilayer MoS₂ FETs is larger than any previously reported values of single to bilayer MoS₂ grown on SiO₂/Si substrate with a SiO₂ gate oxide. Moreover, our few-layer MoS₂ FETs exhibited the highest mobility value ever reported for any MoS₂ FETs with a SiO₂ gate oxide. It is presumed that the high mobility behavior of our film could be attributed to low charged impurities of our film and dielectric screening effect by an interfacial MoO_xSi_y layer. The combined preparation route of RF sputtering and post-deposition annealing process opens up the novel possibility of mass and batch production of MoS₂ film.

Recently, MoS₂ has attracted tremendous interest due to its film thickness scalability, its reducibility from bulk to a monolayer without surface dangling bonds or native oxides, and its promising carrier transport properties^{1,2}. In contrast to graphene, which is intrinsically a semimetal with a zero band-gap, MoS₂ is a semiconductor, which makes it a suitable substrate material for 2-dimensional (2D) field effect transistors (FETs)^{3,4}. From an application point of view, a mass-producible growth technique for large-area, continuous, and high-quality MoS₂ film on dielectrics is a pre-requisite. Micromechanical exfoliation method provides the purest MoS₂ flakes with the highest material quality, the sample size is extremely limited^{2,5}. Several attempts have performed by different groups to satisfy those needs for MoS₂ film⁶. Many research groups also have reported promising growth route of CVD-MoS₂^{7–9}. Sulfurization of molybdenum (Mo)^{10,11} and thermolysis of Mo compounds^{10,12} and (NH₄)₂MoS₄¹³ have attempted previously for preparation MoS₂. MoO₃ and MoCl₅ along with sulfur are common precursors for MoS₂-CVD^{14–17}. Such methods usually yielded multilayer and suffered due to non-uniform film thickness and low carrier mobility^{10,14,15,18,19}. Moreover, the synthesized continuous MoS₂ films via the surface treatment exhibits

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very low carrier mobility ($0.02\text{--}7\text{ cm}^2/\text{Vs}$)^{10,14,15,18,20}. Continuous CVD-MoS₂ films have been demonstrated using MoCl₅ without pre-treatment, but the reported carrier mobility is also very low ($0.003\text{--}0.03\text{ cm}^2/\text{Vs}$)¹⁸. Sanne *et al.*²¹ reported mobility value of $24\text{ cm}^2/\text{Vs}$ and $I_{\text{on}}/I_{\text{off}}$ current ratio exceeding 10^7 for top-gated MoS₂ FETs with high- k gate dielectric on Si₃N₄. Ma *et al.*²² demonstrated the vapor-solid growth of few-layer MoS₂ films on (0001) oriented sapphire. They estimated room temperature mobility of $192\text{ cm}^2/\text{Vs}$ from the space-charge limited transport regime of the film. Laskar *et al.*²³ attained large-area MoS₂ films on (0001) oriented sapphire using sulfurization of e-beam evaporated Mo. They reported field-effect mobility of $\sim 12\text{ cm}^2/\text{Vs}$ using Mott-Guirney law with the carrier density of 10^{16} cm^{-3} . Still, the lack of pristine quality, and wafer-scale synthesis of continuous MoS₂ film on SiO₂ is a challenging issue to be addressed.

Recently, there are few attempts to revive the sputtering technique for the growth of thin MoS₂ film^{24–26}. However, the reported films are either relatively thick or the reported electrical and optical properties are rare and poor^{27–29}. Muratore *et al.*²⁷ and Qin *et al.*²⁸ reported the synthesis of continuous few-layer MoS₂ by sputtering method using a MoS₂ target. Tao *et al.*³⁰ reported MoS₂ film using Mo target sputtered in vaporized sulfur ambient, but the grown MoS₂ film also exhibited p-type behavior with hole mobility up to $\sim 12.2\text{ cm}^2/\text{Vs}$ and low on/off current ratio of $\sim 10^3$.

Herein, we report a simple and mass-scalable approach for thin MoS₂ films via MoS₂-RF sputtering combined with the post-deposition annealing process for the first time. From Raman spectra and photoluminescence (PL), it has been shown that the crystalline quality of the as-sputtered MoS₂ films was highly enhanced through the post-deposition annealing process. Synthesized bilayer MoS₂ films exhibited high field-effect mobility of $\sim 29\text{ cm}^2/\text{Vs}$ and a current on/off ratio of $\sim 10^4$. The mobility increased up to $\sim 173\text{--}181\text{ cm}^2/\text{Vs}$, respectively, for few-layer MoS₂ films. To the best of our knowledge, the mobility value of our bilayer MoS₂ FETs is larger than any reported results of single to bilayer MoS₂ FETs grown on SiO₂/Si with a SiO₂ gate oxide. Furthermore, the mobility value ($\sim 173\text{--}181\text{ cm}^2/\text{Vs}$) of our few-layer MoS₂ FETs is the highest ever for any MoS₂ FETs with a SiO₂ gate oxide. It is much higher than that of single crystal exfoliated MoS₂ flakes on SiO₂/Si substrate³¹ and comparable to the value of bulk MoS₂, room temperature mobility limited by phonon-scattering³².

Results and Discussion

MoS₂ films of different thicknesses were deposited by adjusting RF magnetron sputtering time such as 1, 3, 5 and 15 min onto SiO₂/Si, quartz and sapphire substrates. The substrate temperature was varied from RT to 500 °C. As-sputtered films were subjected to post-deposition annealing treatment at 700 °C in the sulfur and Ar environment to improve their crystallinity. The detailed scheme for preparation and annealing processes is illustrated in Fig. 1(a). Optical microscopy images of sulfurized MoS₂ films at 1, 3 and 5 min. sputtered on SiO₂/Si substrate are shown in Fig. 1b–d.

Raman spectra of the as-sputtered MoS₂ films are shown in Fig. 2a–c. The as-sputtered MoS₂ films exhibit the E_{12g}¹ and A_{1g}¹ mode peaks with low intensity. It might be due to low crystalline quality and the presence of defects contributes to the broad and low intensity of the peaks. The strong substrate related peak is observed at 520 cm^{-1} . As the sputter time increases, the Raman scattering peak intensities are slightly enhanced. Additional peaks at ~ 820 and $\sim 992\text{ cm}^{-1}$ are related to the oxygen bonds and characteristic peaks of MoO₃ (alpha(α)-MoO₃)³³. The symmetric stretch of 820 cm^{-1} (A_g¹, B_g¹) is a terminal Mo=O bond and the 995 cm^{-1} (A_g¹, B_g¹) is an asymmetric stretch of the terminal Mo=O bond along the a- and b-axes^{24,25,34}. MoS₂ films are highly sensitive to moisture and oxidize easily. It has been also proposed that conventional sputter-deposited MoS₂ film contains oxygen substituted for sulfur atoms in the MoS₂ crystal lattice during film growth²⁶.

Figure 2a–c shows that Raman spectra variation through post-deposition annealing. The Raman peak enhancement indicates that the high-temperature annealing in the presence of sulfur and Ar greatly improved the crystallinity of as-sputtered MoS₂ film. Moreover, MoO₃-related peaks were significantly suppressed for the annealed MoS₂ films. Through the post-deposition annealing in sulfur and Ar, the MoO₃ is believed to be transformed into a crystalline MoS₂ structure^{10,35}. For the 1 min-sample (MoS₂ sputtered for 1 min and annealed at 700 °C for 1 hour), the Raman peak difference between E_{12g}¹ and A_{1g}¹ mode is $\sim 20.5\text{ cm}^{-1}$, which is close to that of the exfoliated bilayer MoS₂³⁶. Figure 2d,e shows the Raman spectra according to the different annealing times from 30 min to 3 hours. The peak intensities are increased slightly with increase of annealing time. In order to focus oxygen-related peaks more precisely, the Raman analysis was performed for thick MoS₂ films; as-sputtered films for 15 min at RT and 400 °C, and annealed MoS₂ films (Figure S1). The thick film sputtered at RT exhibited strong MoO₃ peaks at ~ 822 and $\sim 992\text{ cm}^{-1}$ (Figure S1c,d). The oxygen peak intensities were reduced at higher substrate temperature (400 °C), but decreased most through the post-deposition annealing at 700 °C (the as-synthesized film was originally sputtered at RT). Raman mapping was performed over an area of $30\text{ }\mu\text{m} \times 30\text{ }\mu\text{m}$ for 1 min-sample as shown in Fig. 2f–h. The E_{12g}¹ and A_{1g}¹ mode peaks appear at $\sim 384.82\text{--}384.92$ (with a standard deviation 0.048 cm^{-1}) and $\sim 405.19\text{--}405.29\text{ cm}^{-1}$ (with a standard deviation 0.049 cm^{-1}), respectively. The peak difference (Δk) values are in the range of $\sim 20.27\text{--}20.47\text{ cm}^{-1}$ (with a standard deviation 0.066 cm^{-1}), corresponding to the MoS₂ bilayer^{14,36}. For the 3 min-sample (MoS₂ sputtered for 3 min and annealed at 700 °C for 1 hour, Figure S2), E_{12g}¹ and A_{1g}¹ mode are located in the range of $\sim 382.23\text{--}382.33\text{ cm}^{-1}$ (with a standard deviation 0.05 cm^{-1}) and $\sim 407.29\text{--}407.39\text{ cm}^{-1}$ (with a standard deviation 0.045 cm^{-1}), respectively, with Δk values in the range of $\sim 24.96\text{--}25.16\text{ cm}^{-1}$ (with a standard deviation 0.066 cm^{-1}), corresponding to few-layer MoS₂ film³⁶. For the 5 min-sample (MoS₂ sputtered for 5 min and annealed at 700 °C for 1 hour), the E_{12g}¹ mode position downshifted to $\sim 380.63\text{--}380.73\text{ cm}^{-1}$ (with a standard deviation 0.05 cm^{-1}) and the A_{1g}¹ mode upshifted to $\sim 408.29\text{--}408.39\text{ cm}^{-1}$ (with a standard deviation 0.047 cm^{-1}). The Δk value is increased to $\sim 27.56\text{--}27.76\text{ cm}^{-1}$ (with a standard deviation 0.070 cm^{-1}), suggesting that film thickness increment. The Raman measurement was also performed for as-synthesized MoS₂ sputtered at various substrate temperatures from 200 to 500 °C (Figure S3). The as-sputtered film at a substrate temperature of 200 °C exhibits two characteristic MoS₂ Raman peaks with low intensity (E_{12g}¹ mode at $\sim 381\text{ cm}^{-1}$ and A_{1g}¹ mode at $\sim 411\text{ cm}^{-1}$). At higher substrate temperatures of 300, 400

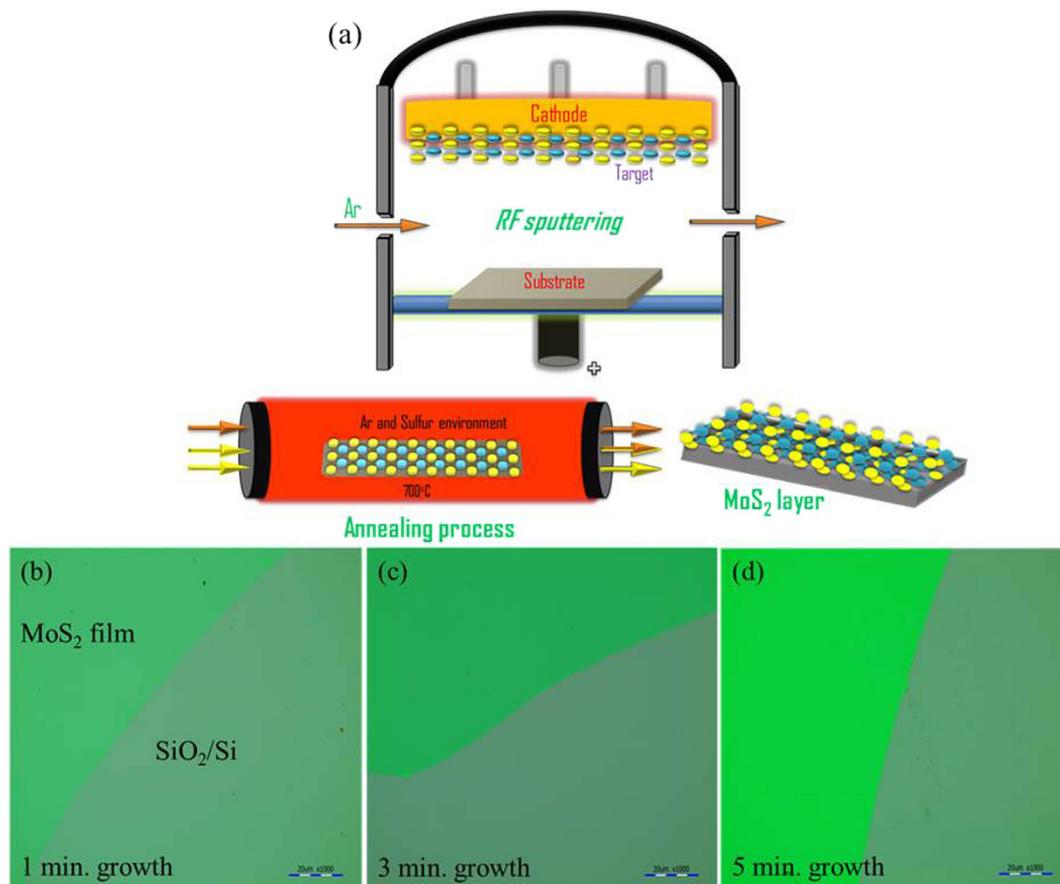


Figure 1. (a) Schematic representation of the experimental set-up. The RF sputtering technique was used to prepare as-sputtered MoS₂ layer. Post-deposition annealing treatment was performed to further enhance crystalline quality in as-sputtered MoS₂ under Ar and sulfur environment. Optical images of MoS₂ films grown on SiO₂/Si substrate. (b) MoS₂ sputtered for 1 min; (c) MoS₂ sputtered for 3 min; and (d) MoS₂ sputtered for 5 min.

and 500 °C, the Raman peak intensities are slightly varied and MoO₃ peaks at ~822 and ~993 cm⁻¹ are reduced. XRD was performed to investigate the structural properties of MoS₂ film. XRD patterns of as-sputtered MoS₂ thin films and the corresponding annealed films are shown in Figure S4a–c. For as-sputtered films, only a silicon substrate-related peak at $2\theta = 33^\circ$ is observed, supporting the amorphous structure of RT-sputtered MoS₂ film. However, (002) lattice oriented diffraction line is observed at $2\theta = 14.2^\circ$ for annealed MoS₂ films. The strong (002) peak is present when the periodicity in c-axis is normal to the MoS₂ film plane which is in good agreement with the previous results^{37,38}. As-sputtered MoS₂ films sputtered at higher substrate temperatures revealed a very weak (002) peak and intensity tends to increase with the increase of sputtering temperature from 200 °C to 500 °C (Figure S5). Thus, Raman and XRD analysis revealed that increase of sputtering temperature improves the film quality and reduces oxygen content but is not sufficient for obtaining high quality MoS₂ film; post-deposition annealing improves film quality the most.

XPS analysis was used to measure binding energies of Mo and S atom. For the 1 min-sample, Mo 3d peaks at 229.1 and 232.2 eV are exhibited (Fig. 3a), which is attributed to the doublet of Mo 3d_{5/2} and Mo 3d_{3/2}, respectively³⁹. Also sulfur atoms-related 2S pathetic peak is observed at 226.3 eV. S²⁻ peaks are also observed (Fig. 3b) at 161.9 and 163.1 eV due to S 2p_{1/2} and S 2p_{3/2}, respectively. In addition, a peak at 235.9 eV corresponds to the Mo⁶⁺ of MoO₃⁴⁰. For the 3 min and 5 min-sample, the observed peaks are slightly shifted to lower binding energies, which may be due to the increment of the number of layers. All these results are in good agreement with the reported values for MoS₂ crystal⁴¹. The intensity of Mo⁶⁺ peaks decreased with increasing growth time. The Mo⁶⁺ peaks indicate that some oxygen is incorporated in the grown MoS₂ film. Oxygen can be incorporated as substitutional atoms at sulfur sites⁴², as atoms bound to Mo atoms at plane edges²⁶, as an intercalant between basal planes as O₂ or moisture (H₂O)⁴³, or as an interfacial Mo-oxide layer due to Mo-oxygen bonding at the MoS₂-SiO₂ interface^{27,28}. XPS survey spectra of Figure S7 show that the total oxygen and silicon signal decreases with increasing sputtering time. This could be explained as the probability of electrons escaping from the SiO₂ substrate reduces exponentially with increasing MoS₂ thickness³¹.

XPS depth profile analysis was performed to investigate the interfacial structure of the MoS₂/SiO₂ film. A 1 keV Ar ion beam was used for sputtering purpose. XPS survey spectra depict that increment of oxygen peak as well as decrement of Mo core level peak with the increase of etching time (Figure S8c). The expanded view of Mo 3d core peak variations are displayed in the Fig. 4a as a function of etching time. Before the sputter etching, the peaks

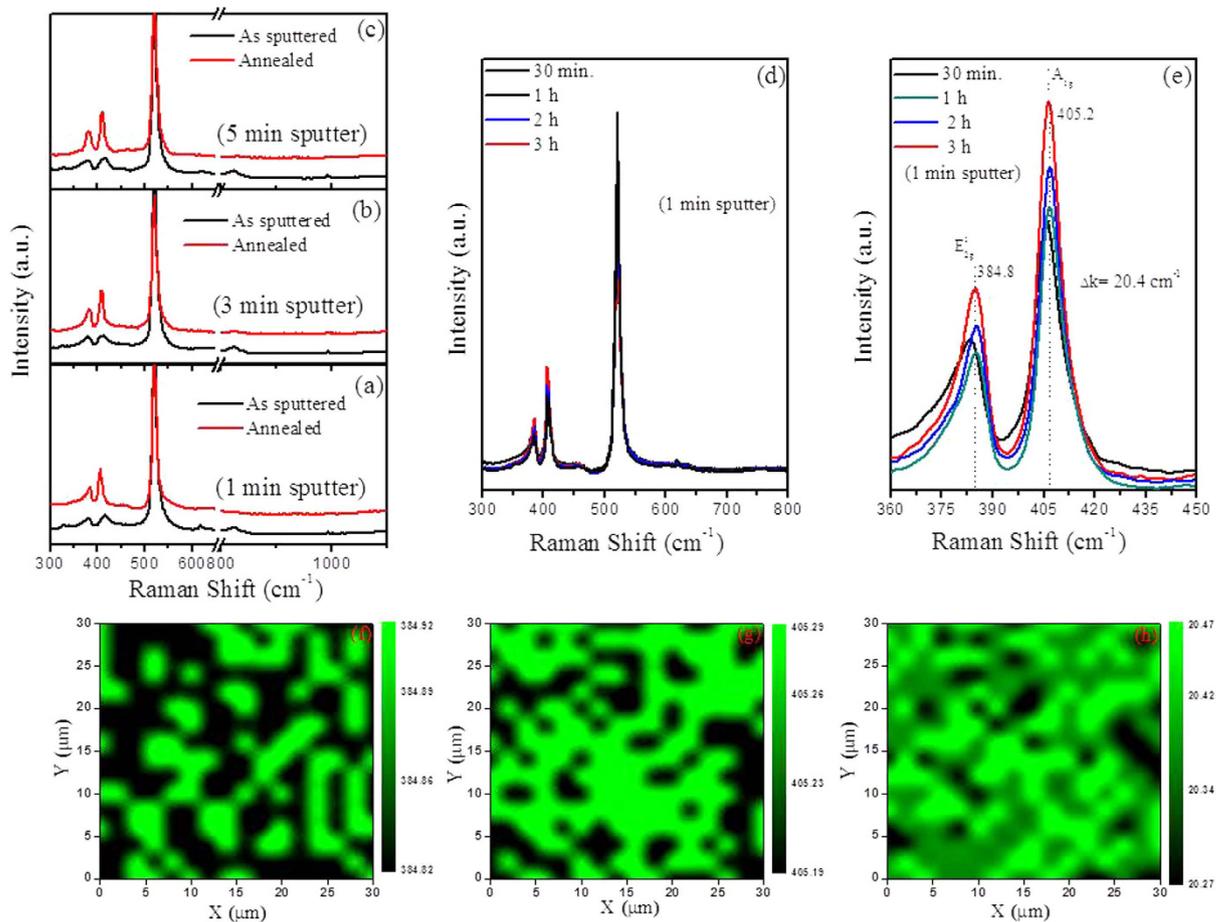


Figure 2. (a–c) Raman spectra of as-sputtered and annealed MoS₂ films; (d) Raman spectra of MoS₂ films annealed at different times of 30 min, 1 hour, 2 hours and 3 hours; (e) Magnified view of Raman spectra of figure (d); (f–h) Raman mapping for 1-min sample (30 μm × 30 μm). (f) E_{2g}¹ mode appears at 384.82–384.92 cm⁻¹ (with a standard deviation 0.048 cm⁻¹) (g) A_{1g} mode appears at 405.19–405.29 cm⁻¹ (with a standard deviation 0.049 cm⁻¹) (h) The measured frequencies difference (Δk) is in the range of 20.27–20.47 cm⁻¹ (with a standard deviation 0.066 cm⁻¹).

of Mo⁴⁺ 3d states are the main part of the spectra, and a small amount of MoO₃ state exists on the surface. When the film is etched by ion beam, there is a chemical shift of its binding energy toward smaller values. The shift is attributed to the change in the chemical states of Mo⁴⁺ from the film surface to inner⁴⁴. The Mo⁶⁺ peak of MoO₃ is highly suppressed after etching for 10 sec. So, the Mo⁶⁺ peaks are mainly originated from the surface oxidation of MoS₂. The peak shift proceeds until 60 sec. After 60 sec, the binding energy shifts back toward higher values. From the Fig. 4b, sulphur related S²⁻ peaks are decreased and broadened as etching proceeds due to the damage induced by Ar etching, and the peaks almost disappear after etching for 50–60 sec (Fig. 4b, Figure S8c, Supporting Information). On the contrary, Mo peaks still exist after 60 sec. Hence, it is highly likely that these Mo could be combined with oxygen atoms or Si atoms in SiO₂ and form as a molybdenum oxide (MoO_x), or molybdenum silicon oxide (MoO_xSi_y) layer. The Si 2p peak in Fig. 4d is exhibited at ~102 eV before Ar etching, and it upshifts towards ~103.2 eV, which is the binding energy of SiO₂. It is suspected that the Si 2p peak at ~102 eV is due to the MoO_x-SiO_x bonding⁴⁵. The Si 2p binding energy at ~102 eV is very close to that of (MoO₃)₇₀(SiO)₃₀ (102.5 eV)⁴⁵.

We later discuss that the interfacial layer can alter the electrical properties of MoS₂ film. The XPS depth profiling was also performed for a very thick MoS₂ film (Figure S8) and observed results are also similar to few-layer MoS₂.

Figure 5a,b shows the cross-sectional high-angle annular dark-field (HAADF) image and the corresponding electron energy loss spectroscopy (EELS) spectra for 5 min-sample. For the position 1 and position 2, 'Si' and 'SiO₂' are detected at ~99 eV and ~105 eV, respectively, and 'O' is detected at ~525 eV. Therefore these two points are clearly SiO₂. A sulfur is detected at ~160 eV from the region 3 and 4, and not from the position 1, 2 and 5, indicating that point 3 and 4 are MoS₂. It is thought that position 2 looks bright due to higher scattering of Mo. The position 5 is an epoxy material exhibiting only C spectrum. The comparison of bright field and HAADF image (Fig. 5c,d) indicates that the region 2 is an interfacial layer of the MoS₂/SiO₂. It is suspected that during sputtering process, Mo adlayers are initially formed at the interface of MoS₂/SiO₂ and the Mo layers diffused into SiO₂ during

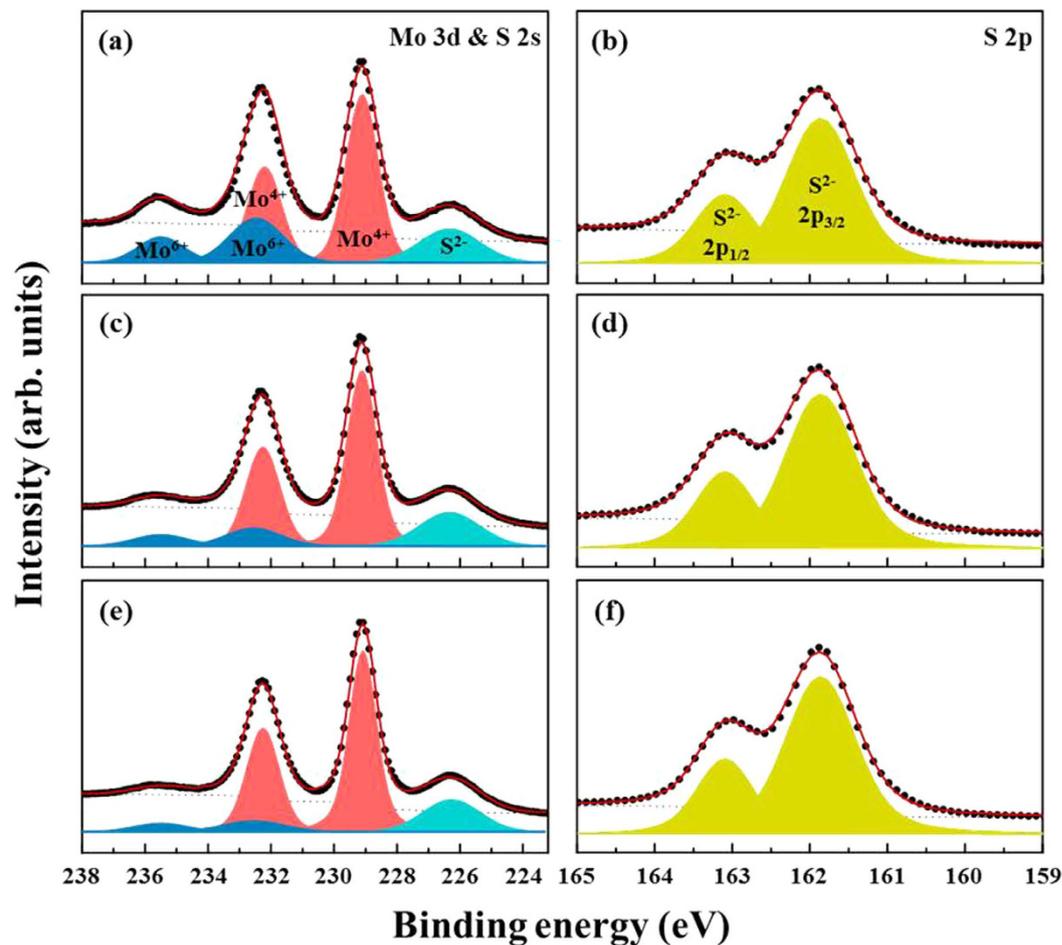


Figure 3. XPS spectra of MoS₂ films annealed at 700 °C. Mo and S atoms binding energy spectra for different sputter time: (a,b) 1 min, (c,d) 3 min, and (e,f) 5 min.

the annealing step, resulting in the formation of MoO_xSi_y layer. The diffused interfacial layer appears brighter due to higher scattering with heavier atoms in that region than that in pure SiO₂ film.

Luminescence properties were studied by PL analysis as shown in Figure S10. The PL peaks are very weak and broad for the as-sputtered films. As sputtering time increases, peak position is shifted to a higher wavelength since the film thickness increases^{46,47}. The luminescence peak intensities are significantly increased for the annealed MoS₂ films (Figure S10b). For the 1 min-sample, the major peak is located at ~662 nm (1.87 eV, A peak) and one minor peak at ~620 nm (2 eV, B peak), which corresponds to a direct excitonic transition at the K point of the Brillouin zone of MoS₂. The energy difference (~0.13 eV) is due to the degeneracy breaking of the valence band, which is in a close agreement with the literature^{48,49}. The measured FWHM value for direct transition of peak A is ~67 meV, which is similar to freely suspended samples of MoS₂ (50–60 meV)⁵⁰ and narrower than that of MoS₂ exfoliated onto SiO₂ (100–150 meV)⁵¹. The emission intensity gradually increases with red shift^{52,53} as increase of annealing time as shown in Figure S10c. This strong luminescence behavior is due to bilayer MoS₂ with a highly crystalline structure and support our earlier observation by Raman and XRD analysis that crystalline quality improvement via annealing at 700 °C.

The thickness of the film was analyzed by AFM as shown in Fig. 6a–c. AFM scan was taken at a corner of the MoS₂ film patterned using photolithography and etching process. For the 1 min-sample, the estimated thickness is ~1.4 nm, which is approximately close to bilayer MoS₂^{18,36} (Fig. 6a). The thickness is ~3.8 nm (~5–6 layers) and ~6 nm (~8–10 layers), for the 3 min and 5 min-samples, respectively. Film continuity and uniformity were explored by AFM topographical 2D images. The surface roughness (R_a , average deviation) values over a scanned area of 5 μm × 5 μm are ~0.18 nm, 0.22 nm, ~0.19 nm for 1, 3, and 5 min as-sputtered MoS₂ films, respectively (Figure S11). 2D topographical images of the annealed films are shown in Fig. 6(d–f). The surface roughness (R_a) values are ~0.25 nm, ~0.35 nm, and ~0.29 nm for 1, 3, and 5 min-sample, respectively. These low roughness values support the highly uniform and continuous MoS₂ films. We believe that a wafer-scale MoS₂ could be produced by optimizing the sputtering time and annealing process.

HRTEM analysis was performed to explore the crystalline structure of MoS₂ film (1 min-sample) as shown in Fig. 7. The lower magnification-HRTEM images are exhibited in Fig. 7a,b for a continuous MoS₂ film on the copper grid. Figure S12a shows the HRTEM image over an area of 39 nm × 30 nm for 1 min-sample. The film shows a continuous film with a hexagonal lattice structure. Several types of Moiré fringes are observed and

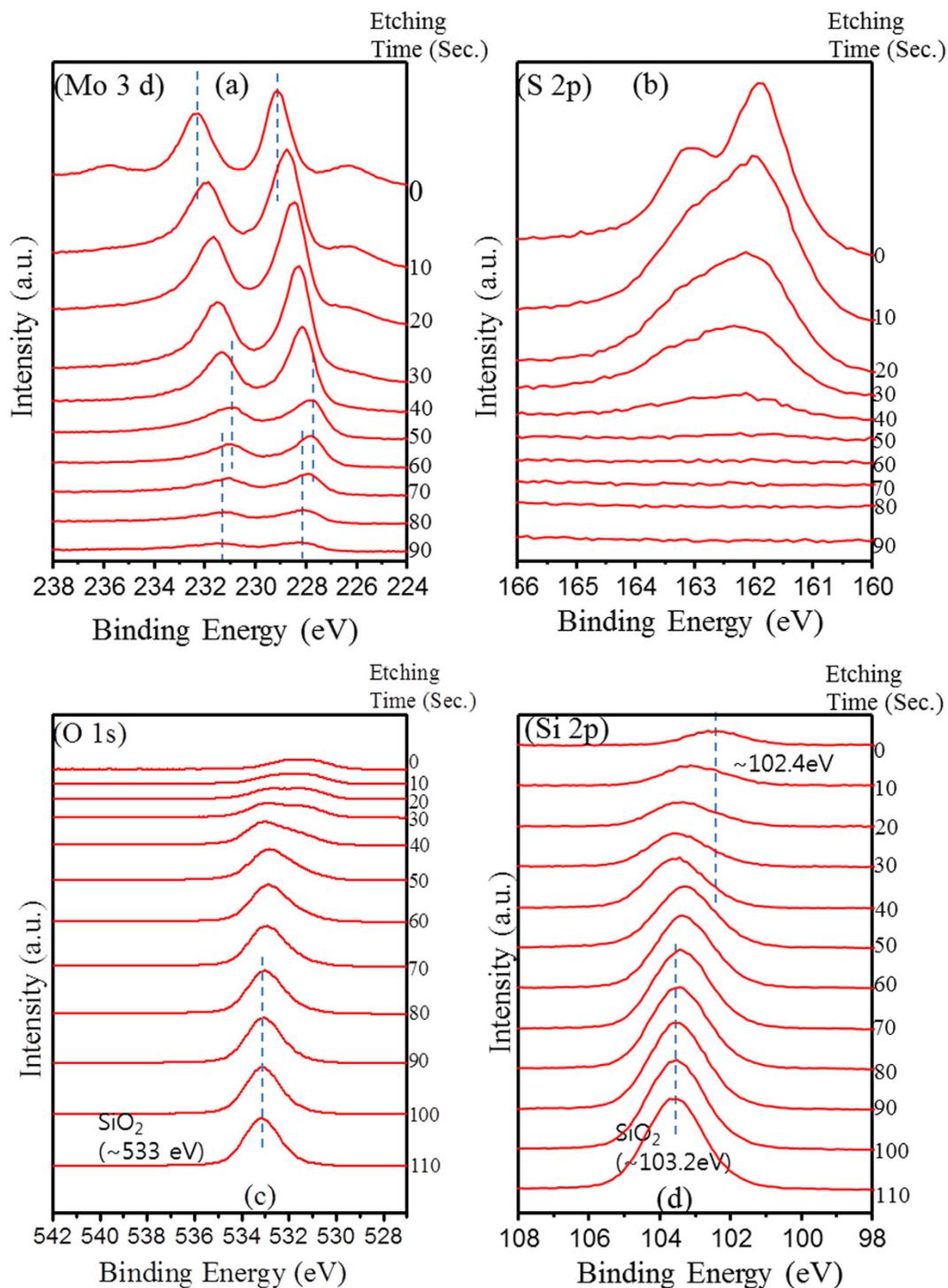


Figure 4. XPS Depth profile of few-layer MoS₂ (5 min-sample). (a) Mo 3d core peaks as a function of etching time. Binding energies at 229.1 and 232.2 eV are associated with Mo⁴⁺ 3d_{5/2} and 3d_{3/2} core levels in MoS₂, respectively, while S 2s appears at 226.3 eV. The peak at 235.9 eV indicates the presence of Mo⁶⁺ (MoO₃) on the surface of the film. (b) Sulfur related S²⁻ peak change with etching time. The sulfur related peaks are eventually disappeared after 50~60 sec. (c,d) O 1s and S 2p peak depth profile with the etching time.

the film consists of mainly bilayer. The observed Moiré fringes in unfolded areas indicate that layers are not Bernal-stacked. A typical Moiré fringes (type B) in Figure S12 were analyzed using fast Fourier transformation (FFT) in Fig. 7d. The exhibited two inverse FFT images (Fig. 7e,f) are extracted from Figure 7c, showing that the two layers are rotated by ~26°. Figure 7g shows a different Moiré pattern (type A in Figure S12) consisting of two layers stacked in a low rotation angle, and the corresponding FFT image is shown in Fig. 7h. The continuous and uniform surface homogeneity was confirmed by FESEM images for 1, 3 and 5-min MoS₂ samples as shown in

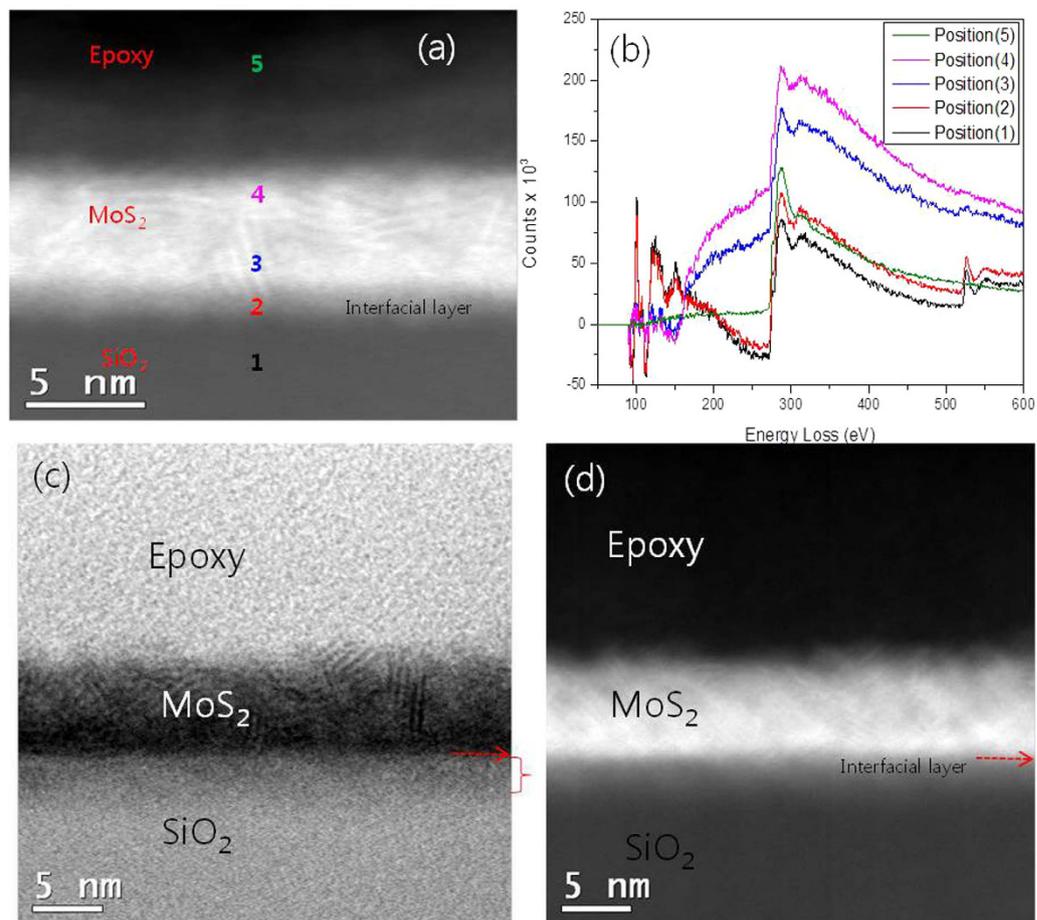


Figure 5. (a) HAADF image for few-layer MoS₂ film (5min-sample) (b) EELS spectra evaluated at different depth positions of film which is labelled as 1,2,3,4 and 5 (c) HRTEM and (d) STEM-HAADF image.

Fig. 7(i–k), respectively. A monolayer is also spotted in Figure S12 (type C). Figure S12b,c shows HRTEM images for the 3 min and 5 min-sample as a supporting information. Large area MoS₂ films with $\sim 1 \times 9$ cm² area and its Raman spectra are shown in Figure S13.

We have fabricated MoS₂ FETs and performed I–V measurement to investigate electrical properties. The schematic diagram of MoS₂ FET structure is given in Figure S15a. The active areas of FETs were defined during the sputtering process using a metal-shadow mask. As-sputtered MoS₂ film exhibited very high resistance in the range between 16 GΩ and 0.2 GΩ. I_d – V_g and I_d – V_d plots of these devices are presented in Figure S14a–f. Our previous results showed that as-sputtered MoS₂ at RT are amorphous structure and are oxidized. As a result, as-sputtered film can exhibit in high channel resistance and low current and mobility^{54,55}. Figure 8a shows that I_d – V_d curves of the 1 min-sample with respect to the back-gate voltages. Figure 8b shows the transfer characteristics of the annealed bilayer MoS₂ FET (1 min-sample). The field-effect mobility was extracted based on the slope of $\Delta I_d/\Delta V_g$ fitted to the linear regime of the transfer curves using the following equation:

$$\mu = \frac{L}{WC_{ox}V_d} \frac{\Delta I_d}{\Delta V_g} \quad (1)$$

where W is the width of the channel (200 μm) L is the length of the channel (2300 μm), C_{ox} is the capacitance per unit area of the gate dielectric (1.15×10^{-8} F/cm²), V_d is the applied drain voltage ($V_d = 1$ V), and $\Delta I_d/\Delta V_g$ is the slope of the linear part of the transfer plot (I_d – V_g), or transconductance. The extracted transconductance, field-effect mobility, and on/off current ratio is $\sim 2.9 \times 10^{-8}$ S, 29 cm²/Vs and $\sim 10^4$, respectively, at $V_d = 1$ V. The linear drain current and the transconductance values at $V_d = 1$ V are displayed in the Figure S15b. The transfer characteristics and I_d – V_d curves for few-layer MoS₂ FETs (3 and 5 min-sample) are shown in Figure S15c,d. The extracted transconductance values are $\sim 1.81 \times 10^{-7}$ S and $\sim 1.73 \times 10^{-7}$ S for the 3 min and 5 min-sample, respectively, which are ~ 6 times greater than bilayer MoS₂ (1 min-sample). The current on/off ratio values are $\sim 2 \times 10^3$ – 4×10^4 for few-layer MoS₂ FETs. The extracted field-effect mobility is ~ 181 and ~ 173 cm²/Vs for 3 min-sample and 5 min-sample, respectively.

Table 1 compares field-effect mobility and I_{on}/I_{off} values of our results with previously reported MoS₂ FETs. A significant enhancement can be noted in our MoS₂ FETs. It is interesting to compare with the recent reported mobility of ~ 12 cm²/Vs for thin MoS₂ film, but the mobility decreased significantly to ~ 0.44 cm²/Vs for

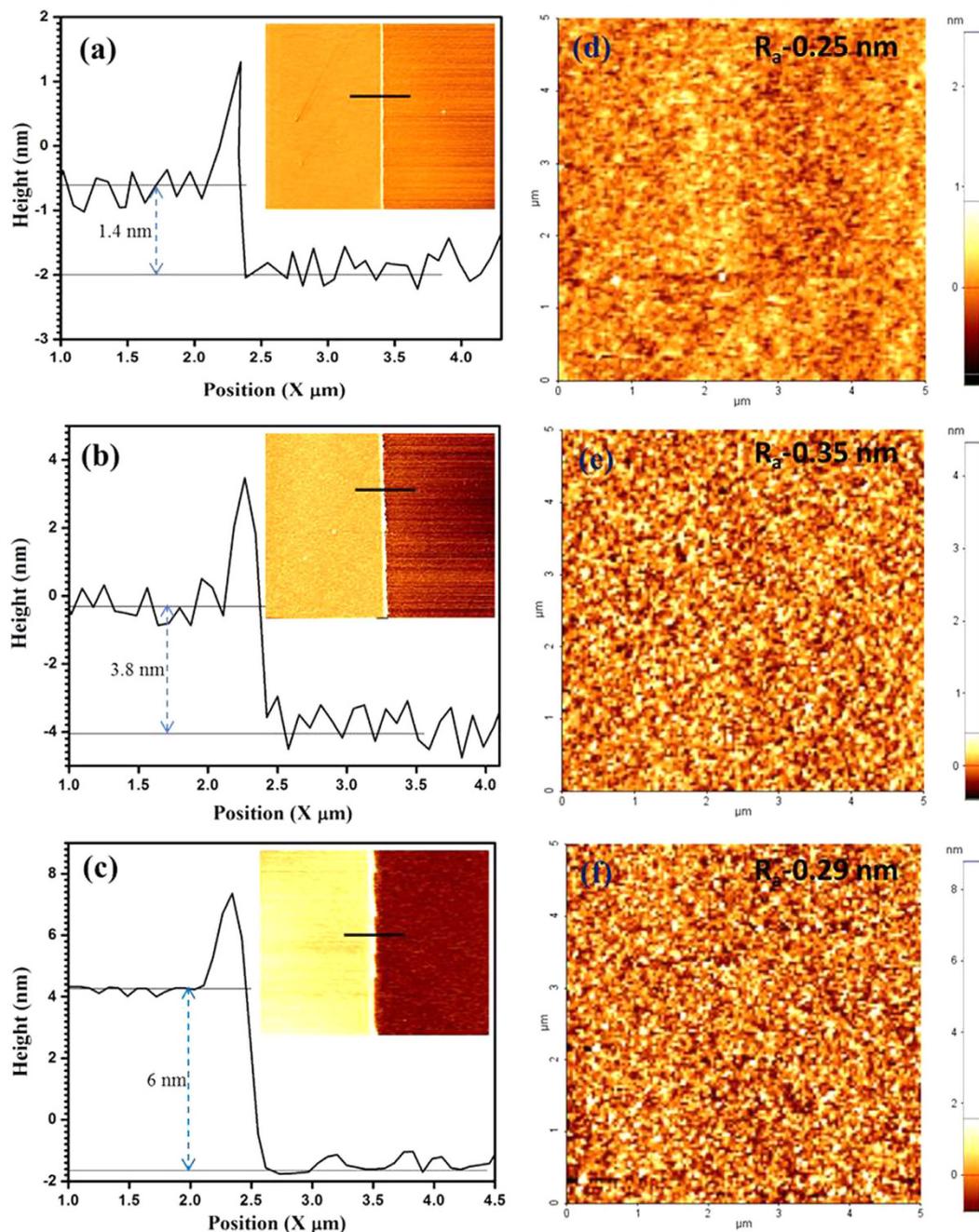


Figure 6. (a–c) AFM height profiles of annealed MoS₂ films sputtered at 1, 3, and 5 min. Inset figure: 2D cross sectional images of the corresponding annealed MoS₂ films; (d–f) Topographical images of annealed MoS₂ films sputtered at 1, 3, and 5 min.

~6.4 nm-MoS₂ due to the incomplete transition of MoS₂ from Mo²⁹. To the best of our knowledge, our bilayer MoS₂ FETs have higher mobility than any of latest results: exfoliated monolayer MoS₂ FETs of 0.1–10 cm²/Vs, 10–15 cm²/Vs for exfoliated bilayer MoS₂², and ~17 cm²/Vs for CVD-grown single crystal bilayer MoS₂⁵⁶. It should be noted that some reports exhibiting very high mobility values for MoS₂ film in Table 1 is due to the substrate effect such as sapphire or high-k gate oxide effect.

Besides, the mobility (173–181 cm²/Vs) of our few-layer MoS₂ is the highest value ever for any MoS₂ FETs with SiO₂ gate dielectrics. Ayari *et al.*³¹ reported 10–50 cm²/Vs of mobility from single crystal exfoliated MoS₂ flakes with 8–40 nm thickness. Our sputtered-MoS₂ films have small grain sizes, which are smaller compared with an exfoliated MoS₂. An important question then remains, what could be the possible mechanism for the high mobility behavior of our MoS₂ film? For current 2D crystal materials, electron mobility is mostly dominated by charged impurity scattering, and the mobility values achieved to date are far below the intrinsic potential in these materials⁵².

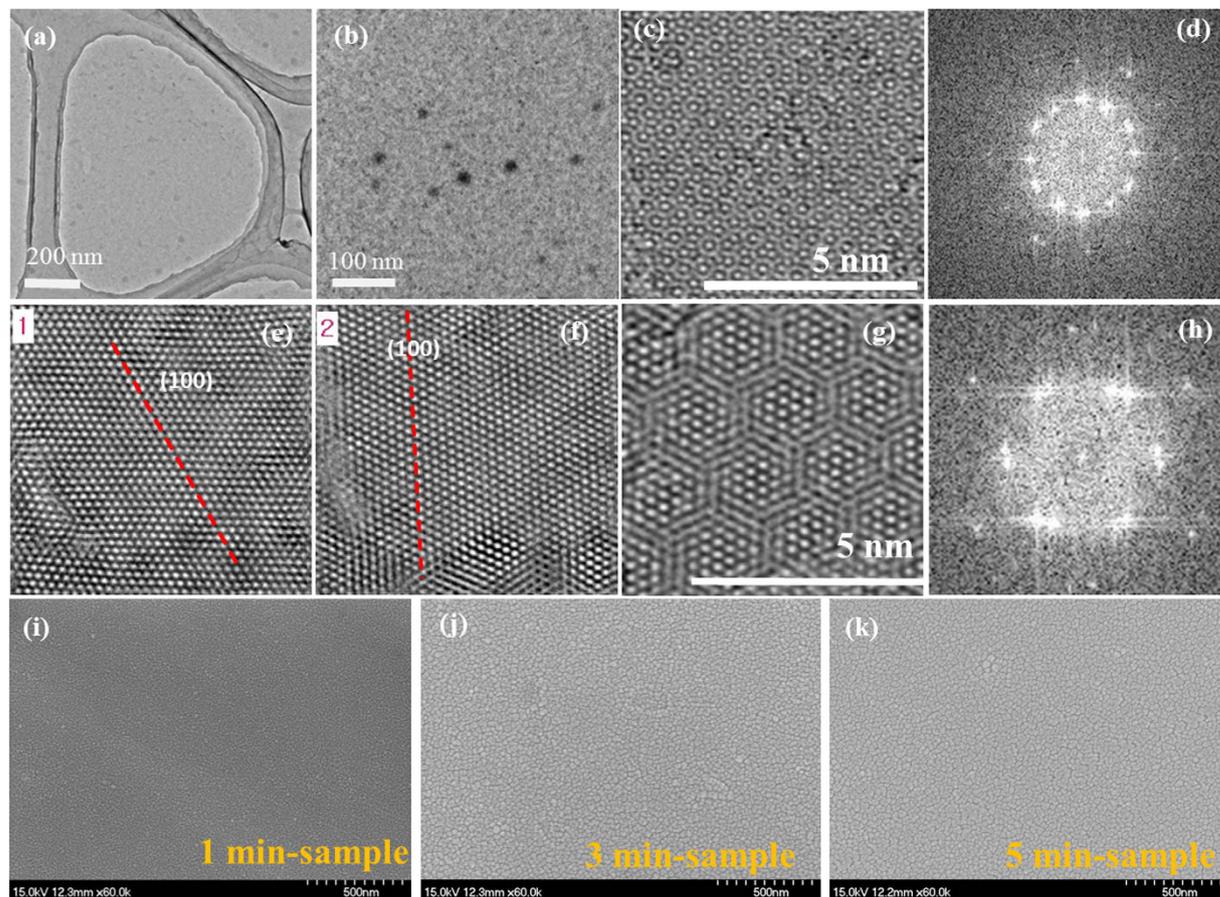


Figure 7. HRTEM images of 1 min-sample. **(a,b)** Low-magnification TEM image; **(c)** Moiré pattern of a bilayer-MoS₂ area; **(d)** Fast Fourier transformation (FFT) image corresponding to the TEM image **(c)** supporting a bilayer MoS₂ film; **(e,f)** Inverse FFT images of **(d)** showing the two layers are not Bernal-stacked, but rotated by ~26°; **(g)** Moiré pattern of a region in which two layers are stacked in a low rotation angle; **(h)** FFT image corresponding to the TEM image **(g)**; **(i-k)** FE-SEM images of annealed MoS₂ films sputtered at 1, 3, and 5 min.

We think that the high mobility behavior of our film could be attributed to low charged impurities of our film and dielectric screening effect by the interfacial MoO_xSi_y layer. In our process, MoS₂ films were directly sputtered on SiO₂/Si substrate at high vacuum and transistors were fabricated without transfer step, while the conventional CVD-grown MoS₂, except exfoliated MoS₂, usually needs the wet-transfer process onto a desired dielectric substrate and it make high contamination. Since sputtering process is performed in a high vacuum chamber, the chemical residues and gaseous adsorbates could be minimized. In addition, the dielectric surface dangling bonds could be also minimized due to a strong interaction of Mo and O on SiO₂ of the interfacial layer. Thus, low charged impurities could reduce the Coulomb scattering, resulting in high mobility values in the sputtered-MoS₂⁵³.

It is also well known that a bulk α -MoO₃ possesses very high relative dielectric constants (>500 for α -MoO₃)⁵⁷. And the dielectric constants of an atomically thin α -MoO₃ is still high even though it is low compared with its bulk value⁵⁸. Thus, the MoO_xSi_y could reduce Coulomb scattering effects due to its high-k value as well as low dielectric dangling bonds. We have also prepared MoO₃ film on SiO₂/Si substrate via a reactive sputtering using Mo target. XPS data of the sulfurized MoS₂ from Mo target also have the MoO₃ peak similar to the previous results (Figure S16). The as-sputtered MoO₃ exhibited very high resistance due to a wide bandgap of the material. On the other hand, the sulfurized few-layer MoS₂ FETs (from MoO₃) exhibited high mobility values (~44 cm²/Vs) (Figure S17). This experiment also supports our hypothesis. The fact that few-layer MoS₂ has much higher mobility value than that of bilayer MoS₂ reflects a critical role of Coulomb interaction distance upon the mobility values since thicker film has longer interaction distance. We compared hysteresis in transfer curves of FETs made by exfoliated-MoS₂, CVD-grown MoS₂, and sputtered-MoS₂ (Figures S18 and S19). It is well known that the origin of hysteresis of conventional FETs is due to the trapping and detrapping of carriers⁵⁹. The trapping and detrapping can occur at the interface of the MoS₂/SiO₂ or at the top surface of MoS₂. Imperfect interface between MoS₂ and SiO₂ such as foreign molecules trapped at the interface or dielectric dangling bonds could contribute to the interface trap of MoS₂/SiO₂. Chemical residues or moisture or oxygen on the MoS₂ surface could contribute the charge trapping at the top surface of MoS₂ film. Water and oxygen in ambient environment also have been reported to cause hysteresis of MoS₂ FETs due to the charge transferring on MoS₂ top surface⁵⁹.

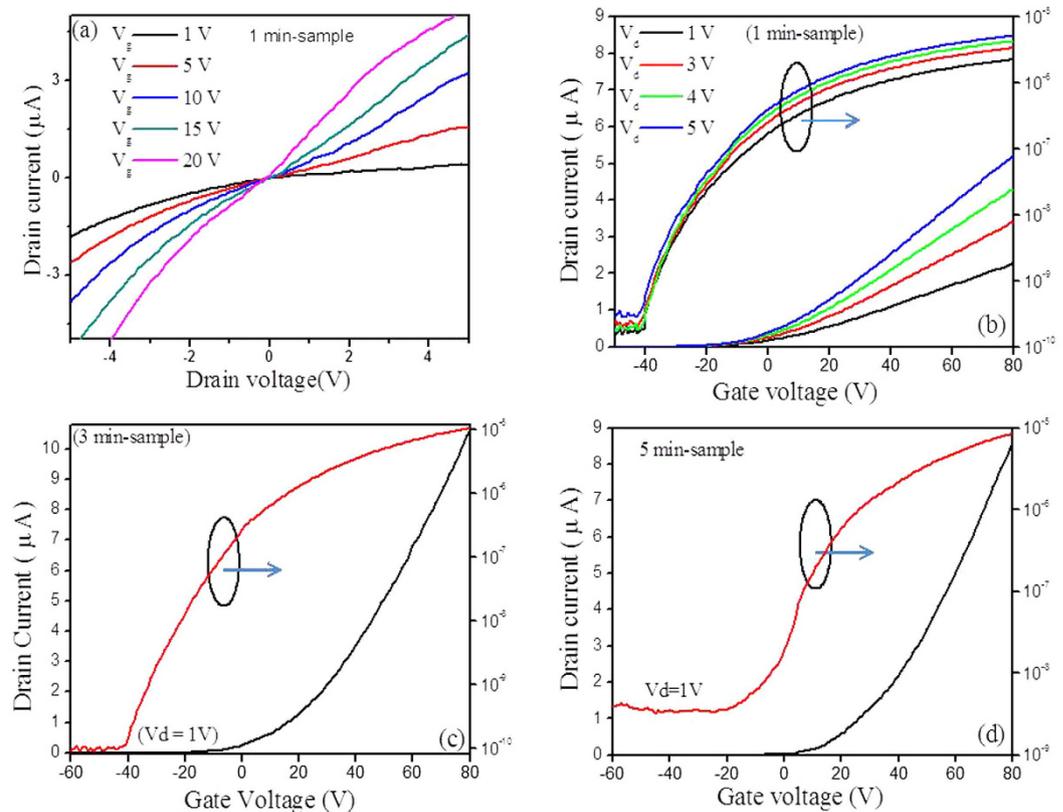


Figure 8. (a) I_d - V_d of MoS₂ FET of 1-min sample; (b) I_d - V_g of MoS₂ FET (1 min-sample) at $V_d = 1, 3, 4$ and 5 V; (c,d) I_d - V_g of MoS₂ FETs, 3 min-sample (c), and 5 min-sample (d) at fixed $V_d = 1$ V.

We compared the hysteresis under vacuum environment to prevent such extrinsic and environmental effects and focus on the trapping at the MoS₂/SiO₂ interface⁶⁰. The exfoliated-MoS₂ and CVD-grown MoS₂ exhibited large hysteresis in their I_d - V_g curves. On the contrary, the sputtered-MoS₂ film exhibited small hysteresis. Such improvement in the hysteresis can be attributed to the small trap at the MoS₂/SiO₂ interface of the sputtered-MoS₂ film. It is thought that charge scattering due to charge trapping is reduced due to the interfacial layer and enhance the mobility behavior of our sputtered-MoS₂ film.

Conclusions

We have successfully demonstrated the growth of large-area and continuous bilayer to few-layer MoS₂ on SiO₂/Si substrate via RF sputtering combined with the post-deposition annealing process. The crystalline quality of the as-sputtered films was substantially improved via annealing at 700 °C in the sulfur and argon environment. The bilayer MoS₂ FETs exhibited a high field-effect mobility of ~ 29 cm²/Vs and an on/off ratio of $\sim 10^4$. The mobility value of our bilayer MoS₂ FETs is larger than any of latest results of single to bilayer MoS₂ grown on a SiO₂/Si substrate with a SiO₂ gate oxide. The mobility for few-layer MoS₂ FETs increased to ~ 173 – 181 cm²/Vs. Our few-layer MoS₂ FETs exhibited the highest mobility value ever for any MoS₂ FETs with a SiO₂ gate oxide. It is presumed that the high mobility behavior of our film could be attributed to low charged impurities of our film and dielectric screening effect by the interfacial MoO_xSi_y layer. The combined synthesis route of MoS₂-RF sputtering with the post-deposition annealing process could open up the possibility of mass and batch production of MoS₂ film. We believe our proposed strategy will pave the way for applications of MoS₂ in future electronics and optoelectronics.

Method

The various sizes of SiO₂ (300 nm)/Si substrates ranging from 1×1 cm² to 3×3 cm² were used for the film preparation process. All the substrates were cleaned in acetone, methanol, isopropyl alcohol (IPA) solution and deionized (DI) water and then dried and baked for 5 min. After loading the SiO₂/Si substrates into a sputtering chamber, the chamber was vacuumed at 1×10^{-6} Torr. Before the deposition process, the MoS₂ target (99.99% purity) was pre-sputtered in a pure argon (Ar) atmosphere for 5 min in order to remove the oxide layer on the surface of the target. The MoS₂ films were sputtered at various temperatures: RT, 200, 300, 400 and 500 °C. The chamber pressure was maintained at 10 mTorr during the deposition in an Ar atmosphere, and the RF power was kept constant at 25 W for 1 min. The temperature variation in the chamber was monitored through a thermocouple. The as-sputtered MoS₂ films were post-annealed at 700 °C under Ar and sulfur environment to improve the crystalline quality of the films. The as-deposited films were placed in an annealing chamber and heated up

S. No.	Growth method	I_{on}/I_{off}	Mobility [cm^2/Vs]	Ref.
1	Sputtering (MoS_2) + CVD	$\sim 10^4$	~ 29 (~ 1.4 nm) ~ 173 – 181 (~ 3.8 – 6 nm)	This work
2	CVD (MoO_3 + S)	$\sim 10^7$	24	Appl. Phys. Lett., 106 (2015) 062101
3	Sputtering (MoS_2)	$\sim 10^3$	12.2	Nanoscale, 2015,7, 2497–2503
4	MoO_3 powder + Mo substrate+ CVD		192	Appl. Phys. Lett., 105 (2014) 072105
5	Ebeam (Mo) + CVD		12 ± 2	Appl. Phys. Lett., 102 (2013) 252108.
6	Sputtering (Mo) + CVD	$\sim 1.5 \times 10^6$ – 5×10^4	12 (~ 1.1 nm) 0.44 (~ 6.4 nm)	ACS Appl. Mater. Interfaces, 2014, 6 (23), 21215–21222
7	CVD (MoCl_5 + S)	$\sim 10^4$ – 10^5	0.003–0.03	Scientific Reports 3 : 1866 DOI: 10.1038/srep01866
8	CVD (MoO_3 + S) on rGO	$\sim 10^4$	0.02	Adv. Mater. 2012, 24, 2320–2325
9	CVD (MoO_3 + S)	$\sim 10^6$	2–7	ACS Nano, 2014, 8 (6), 6024–6030
10	Ebeam (Mo) + CVD		0.004–0.04	Small 2012, 8, 966.
11	CVD (MoO_3 + S)	$\sim 10^8$	17	Appl. Phys. Lett. 100, 123104 (2012)
12	CVD (MoO_3 + S)	$\sim 10^4$ – $\sim 10^6$	0.1–0.7	J. Amer. Chem. Society 2013, 135, 5304.
13	CVD (MoO_3 + S)	$\sim 10^3$	0.09	Nano Research 2014, 7 (12) : 1759–1768
14	Thermal (MoO_3) + CVD on sapphire	$\sim 10^5$	~ 0.8	Nanoscale, 2012,4, 6637–6641
15	Thermolysis of $(\text{NH}_4)_2\text{MoS}_4$	$\sim 10^5$	4.7–6	Nano Lett., 2012, 12 (3), 1538–1544
16	Exfoliated (electrochemical)	$\sim 10^6$	1.2	ACS Nano, 2014, 8 (7), 6902–6910
17	CVD (H_2S + Mo)	$\sim 10^5$	0.12	Nanoscale, 2014,6, 2821–2826
18	$\text{Mo}(\text{CO})_6$ + $(\text{C}_2\text{H}_5)_2\text{S}$	$\sim 10^4$	30	Nature, 2015, 520, 656–660
19	CVD (MoO_3 + S)	$\sim 10^6$	3.6 (1L), 8.2 (2L), 15.6 (3L)	Nanoscale, 2015,7, 1688–1695
20	CVD (MoO_3 + S)	10^5 – 10^7	~ 3 to 4	Nat. Mater., 2013, 12, 554–561.

Table 1. Literature values of room temperature field-effect mobility for MoS_2 FETs grown by various methods.

to 700 °C for 30 min, 1 hour, 2 hours, and 3 hours. The carrier gas flow rate was maintained at 100 sccm, and the pressure of chamber was kept at 2×10^{-2} Torr.

Fabrication of the MoS_2 FET devices. The active area of MoS_2 FET was formed during sputtering using a shadow mask. This kind of shadow mask is to avoid any chemical contamination by traditional active area preparation route of photolithography or electron-beam lithography. The metal contacts of 6 nm-Ti/30 nm-Au were prepared by evaporation. After making the electrode contacts, the devices were annealed at 200 °C for 2 hour in a vacuum tube furnace with 100 sccm Ar flow. After the annealing, the resistance of devices decreased significantly. The electrical properties of the fabricated MoS_2 transistors were measured using the 2 probe method at room temperature in a vacuum chamber to avoid oxidation.

Characterization details of MoS_2 films. Synthesized MoS_2 films were analyzed by Raman spectroscopy (Renishaw invia RE04, 512 nm Ar laser) with a spot size of 1 μm and a scan speed of 30 seconds. A Si substrate with a Raman peak of 520 cm^{-1} was used for calibration. X-ray photoelectron spectroscopy (XPS) (PHI 5000 Versa Probe, 25W Al $K\alpha$, 6.7×10^{-8} Pa) and photoluminescence (PL) with a 512 nm wavelength was used. Laser radiation of PL was focused onto the MoS_2 film with a spot-size of around 1 μm . FE-SEM (HITACHI S-4700) and atomic force microscopy (AFM) (Veeco Dimension 3100) were used to check the morphology and thickness of the films. TEM samples were prepared using lacey-carbon Cu grid. The atomic structure of MoS_2 thin films was characterized by a JEOL-2010F TEM with an accelerating voltage of 200 keV. Image acquisition and processing (FFT, IFFT, etc.) were performed using the Gatan Digital Micrograph software (Gatan Microscopy Suite 2.0). The crystallinity of the film was characterized by in-plan X-ray diffraction (XRD, Rigaku) with $\text{Cu-K}\alpha$ radiation operated at 50 KV and 300 mA.

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Author Contributions

S.H. and J.S. initiated the study, performed the extensive experiments and wrote the paper with assistance from the co-authors. D.V. and D.-C.C. analyzed the data. A.K.S., M.Z.I. and M.F.K. help us in electrical transport properties. P.K. did us PL measurement. WS and K.-S.A. performed XPS and XPS depth profile analyses. J.E., W.-G.L. and J.J. Participation included planning, experimental work and discussion. All authors read and approved the final manuscript.

Additional Information

Supplementary information accompanies this paper at <http://www.nature.com/srep>

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