Large-area, continuous and high electrical performances of bilayer to few layers MoS$_2$ fabricated by RF sputtering via post-deposition annealing method

Sajjad Hussain$^{1,2}$, Jai Singh$^3$, Dhanasekaran Vikraman$^4$, Arun Kumar Singh$^5$, Muhammad Zahir Iqbal$^5$,†, Muhammad Farooq Khan$^2$, Pushpendra Kumar$^6$, Dong-Chul Choi$^{1,2}$, Wooseok Song$^7$, Ki-Seok An$^7$, Jonghwa Eom$^5$, Wan-Gyu Lee$^8$ & Jongwan Jung$^{1,2}$

We report a simple and mass-scalable approach for thin MoS$_2$ films via RF sputtering combined with the post-deposition annealing process. We have prepared as-sputtered film using a MoS$_2$ 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 MoS$_2$ film. The mobility value of ~29 cm$^2$/Vs and current on/off ratio on the order of ~10$^4$ were obtained for bilayer MoS$_2$. The mobility increased up to ~173–181 cm$^2$/Vs, respectively, for few-layer MoS$_2$. The mobility of our bilayer MoS$_2$ FETs is larger than any previously reported values of single to bilayer MoS$_2$ grown on SiO$_2$/Si substrate with a SiO$_2$ gate oxide. Moreover, our few-layer MoS$_2$ FETs exhibited the highest mobility value ever reported for any MoS$_2$ FETs with a SiO$_2$ 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$_x$Si$_y$ layer. The combined preparation route of RF sputtering and post-deposition annealing process opens up the novel possibility of mass and batch production of MoS$_2$ film.

Recently, MoS$_2$ 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, MoS$_2$ 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 MoS$_2$ film on dielectrics is a pre-requisite. Micromechanical exfoliation method provides the purest MoS$_2$ flakes with the highest material quality, the sample size is extremely limited$^{2,5}$. Several attempts have been performed by different groups to satisfy those needs for MoS$_2$ film$^6$. Many research groups also have reported promising growth route of CVD-MoS$_2$$^7$–$^9$. Sulfurization of molybdenum (Mo)$^{10,11}$ and thermolysis of Mo compounds$^{10,12}$ and (NH$_4$)$_2$MoS$_4$ have attempted previously for preparation MoS$_2$, MoO$_x$, and MoCl$_5$ along with sulfur are common precursors for MoS$_2$-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 MoS$_2$ films via the surface treatment exhibits...
very low carrier mobility (0.02–7 cm²/Vs)\(^{10,14,15,18,20}\). Continuous CVD-MoS\(_2\) films have been demonstrated using MoCl\(_5\) without pre-treatment, but the reported carrier mobility is also very low (0.003–0.03 cm²/Vs)\(^{18}\). Sanne et al.\(^{21}\) reported mobility value of 24 cm²/Vs and \(I_{off}/I_{on}\) current ratio exceeding 10\(^4\) for top-gated MoS\(_2\) FETs with high-k gate dielectric on Si\(_3\)N\(_4\). Ma et al.\(^{22}\) demonstrated the vapor-solid growth of few-layer MoS\(_2\) films on (0001) oriented sapphire. They estimated room temperature mobility of 192 cm²/Vs from the space-charge limited transport regime of the film. Laskar et al.\(^{23}\) attained large-area MoS\(_2\) films on (0001) oriented sapphire using sulfurization of e-beam evaporated Mo. They reported field-effect mobility of ~12 cm²/Vs using Mott-Guiney law with the carrier density of 10\(^{16}\) cm\(^{-3}\). Still, the lack of pristine quality, and wafer-scale synthesis of continuous MoS\(_2\) film on SiO\(_2\) is a challenging issue to be addressed.

Recently, there are few attempts to revive the sputtering technique for the growth of thin MoS\(_2\) 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 MoS\(_2\) by sputtering method using a MoS\(_2\) target. Tao et al.\(^{30}\) reported MoS\(_2\) film using Mo target sputtered in vaporized sulfur ambient, but the grown MoS\(_2\) film also exhibited p-type behavior with hole mobility up to ~12.2 cm²/Vs and low on/off current ratio of ~10\(^5\).

Herein, we report a simple and mass-scalable approach for thin MoS\(_2\) films via Mo-S\(_2\)-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 MoS\(_2\) films was highly enhanced through the post-deposition annealing process. Synthesized bilayer MoS\(_2\) films exhibited high field-effect mobility of ~29 cm²/Vs and a current on/off ratio of ~10\(^5\). The mobility increased up to ~173–181 cm²/Vs, respectively, for few-layer MoS\(_2\) films. To the best of our knowledge, the mobility value of our bilayer MoS\(_2\) FETs is larger than any reported results of single to bilayer MoS\(_2\) FETs grown on SiO\(_2\)/Si with a SiO\(_2\) gate oxide. Furthermore, the mobility value (~173–181 cm²/Vs) of our few-layer MoS\(_2\) FETs is the highest ever for any MoS\(_2\) FETs with a SiO\(_2\) gate oxide. It is much higher than that of single crystal exfoliated MoS\(_2\) flakes on SiO\(_2\)/Si substrate\(^{31}\) and comparable to the value of bulk MoS\(_2\), room temperature mobility limited by phonon-scattering\(^{32}\).

**Results and Discussion**

MoS\(_2\) films of different thicknesses were deposited by adjusting RF magnetron sputtering time such as 1, 3, 5 and 15 min onto SiO\(_2\)/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 MoS\(_2\) films at 1, 3 and 5 min sputtered on SiO\(_2\)/Si substrate are shown in Fig. 1b–d.

Raman spectra of the as-sputtered MoS\(_2\) films are shown in Fig. 2a–c. The as-sputtered MoS\(_2\) films exhibit the E\(_{2g}\) and A\(_{1g}\) 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 ~992 cm\(^{-1}\) are related to the oxygen bonds and characteristic peaks of MoO\(_3\) (alpha(\(\alpha\))-MoO\(_3\))\(^{33}\). The symmetric stretch of 820 cm\(^{-1}\)(A\(_{g}\), B\(_{1g}\)) is a terminal Mo–O bond and the 995 cm\(^{-1}\)(A\(_{g}\), B\(_{1g}\)) is an asymmetric stretch of the terminal Mo–O bond along the a- and b-axes.\(^{24,25,34}\) MoS\(_2\) films are highly sensitive to moisture and oxidize easily. It has been also proposed that conventional sputter-deposited MoS\(_2\) film contains oxygen substituted for sulfur atoms in the MoS\(_2\) crystal lattice during film growth\(^{26}\).

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 MoS\(_2\) films. Moreover, Mo-S\(_2\)-related peaks were significantly suppressed for the annealed MoS\(_2\) films. Through the post-deposition annealing in sulfur and Ar, the MoO\(_3\) is believed to be transformed into a crystalline MoS\(_2\) structure\(^{16,35}\). For the 3 min-sample (MoS\(_2\) sputtered for 3 min and annealed at 700 °C for 1 hour, Figure S2), E\(_{2g}\) and A\(_{1g}\) mode peaks appear at ~384.82–384.92 (with a standard deviation 0.049 cm\(^{-1}\)) and ~411 cm\(^{-1}\), respectively, with E\(_{2g}\) mode position downshifted to ~382.23–382.33 cm\(^{-1}\) (with a standard deviation 0.066 cm\(^{-1}\)). The peak intensities of E\(_{2g}\) and A\(_{1g}\) mode are located in the range of ~382.23–382.33 cm\(^{-1}\) (with a standard deviation 0.05 cm\(^{-1}\)) and ~407.29–407.39 cm\(^{-1}\) (with a standard deviation 0.045 cm\(^{-1}\)), respectively, with \(\Delta k\) values in the range of ~24.96–25.16 cm\(^{-1}\) (with a standard deviation 0.048 cm\(^{-1}\)) and ~405.19–405.29 cm\(^{-1}\) (with a standard deviation 0.049 cm\(^{-1}\)), respectively. The peak difference (\(\Delta k\)) values are in the range of ~20.27–20.47 cm\(^{-1}\) (with a standard deviation 0.066 cm\(^{-1}\)), corresponding to the MoS\(_2\) bilayer. For the 5 min-sample (MoS\(_2\) sputtered for 5 min and annealed at 700 °C for 1 hour, Figure S2), E\(_{2g}\) and A\(_{1g}\) mode peaks appear at ~388.54–388.63 (with a standard deviation 0.048 cm\(^{-1}\)) and ~411.02–411.06 cm\(^{-1}\) (with a standard deviation 0.049 cm\(^{-1}\)), respectively. The + values are in the range of ~24.96–25.16 cm\(^{-1}\) (with a standard deviation 0.048 cm\(^{-1}\)) and ~405.19–405.29 cm\(^{-1}\) (with a standard deviation 0.049 cm\(^{-1}\)), respectively. The peak difference (\(\Delta k\)) values are in the range of ~20.27–20.47 cm\(^{-1}\) (with a standard deviation 0.066 cm\(^{-1}\)), corresponding to the MoS\(_2\) bilayer. For the 3 min-sample (MoS\(_2\) sputtered for 3 min and annealed at 700 °C for 1 hour, Figure S2), E\(_{2g}\) and A\(_{1g}\) mode peaks appear at ~382.23–382.33 cm\(^{-1}\) (with a standard deviation 0.05 cm\(^{-1}\)) and ~407.29–407.39 cm\(^{-1}\) (with a standard deviation 0.045 cm\(^{-1}\)), respectively, with \(\Delta k\) values in the range of ~24.96–25.16 cm\(^{-1}\) (with a standard deviation 0.048 cm\(^{-1}\)) and ~405.19–405.29 cm\(^{-1}\) (with a standard deviation 0.049 cm\(^{-1}\)), respectively. The peak difference (\(\Delta k\)) values are in the range of ~20.27–20.47 cm\(^{-1}\) (with a standard deviation 0.066 cm\(^{-1}\)), corresponding to the MoS\(_2\) bilayer. For the 5 min-sample (MoS\(_2\) sputtered for 5 min and annealed at 700 °C for 1 hour, Figure S2), E\(_{2g}\) mode position downshifted to ~380.63–380.73 cm\(^{-1}\) (with a standard deviation 0.05 cm\(^{-1}\)) and \(\Delta k\) value is increased to ~408.29–408.39 cm\(^{-1}\) (with a standard deviation 0.047 cm\(^{-1}\)). The + value is increased to ~27.56–27.76 cm\(^{-1}\) (with a standard deviation 0.070 cm\(^{-1}\)), suggesting that film thickness increment. The Raman measurement was also performed for as-synthesized MoS\(_2\) 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 MoS\(_2\) Raman peaks with low intensity (E\(_{2g}\) mode at ~381 cm\(^{-1}\) and A\(_{1g}\) mode at ~411 cm\(^{-1}\)). At higher substrate temperatures of 300, 400
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° is observed, supporting the amorphous structure of RT-sputtered MoS$_2$ film. However, (002) lattice oriented diffraction line is observed at 2$\theta$ = 14.2° for annealed MoS$_2$ films. The strong (002) peak is present when the periodicity in c-axis is normal to the MoS$_2$ film plane which is in good agreement with the previous results$^{37,38}$. As-sputtered MoS$_2$ 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 MoS$_2$ 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$_{3/2}$, respectively$^{39}$. Also sulfur atoms-related 2S pathetic peak is observed at 226.3 eV. S$^{2-}$ 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 Mo$^{6+}$ of MoO$_3$.$^{40}$ 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 MoS$_2$ crystal$^{41}$. The intensity of Mo$^{6+}$ peaks decreased with increasing growth time. The Mo$^{6+}$ peaks indicate that some oxygen is incorporated in the grown MoS$_2$ film. Oxygen can be incorporated as substitutional atoms at sulfur sites$^{42}$, as atoms bound to Mo atoms at plane edges$^{26}$, as an intercalant between basal planes as O$_2$ or moisture (H$_2$O)$^{43}$, or as an interfacial Mo-oxide layer due to Mo-oxygen bonding at the MoS$_2$-SiO$_2$ interface$^{27,28}$. XPS survey spectra of Figure S7 show that the total oxygen and silicon signal decreases with increasing MoS$_2$ thickness$^{31}$. XPS depth profile analysis was performed to investigate the interfacial structure of the MoS$_2$/SiO$_2$ film. A 1keV 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

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**Figure 1.** (a) Schematic representation of the experimental set-up. The RF sputtering technique was used to prepare as-sputtered MoS$_2$ layer. Post-deposition annealing treatment was performed to further enhance crystalline quality in as-sputtered MoS$_2$ under Ar and sulfur environment. Optical images of MoS$_2$ films grown on SiO$_2$/Si substrate. (b) MoS$_2$ sputtered for 1 min; (c) MoS$_2$ sputtered for 3 min; and (d) MoS$_2$ sputtered for 5 min.
of Mo\textsuperscript{4+} 3d states are the main part of the spectra, and a small amount of MoO\textsubscript{3} 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 Mo\textsuperscript{4+} from the film surface to inner\textsuperscript{44}. The Mo\textsuperscript{6+} peak of MoO\textsubscript{3} is highly suppressed after etching for 10 sec. So, the Mo\textsuperscript{6+} peaks are mainly originated from the surface oxidation of MoS\textsubscript{2}. The peak shift proceeds until 60 sec. After 60 sec, the binding energy shifts back toward higher values. From the Fig. 4b, sulphur related S\textsuperscript{2−} 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 SiO\textsubscript{2} and form as a molybdenum oxide (MoO\textsubscript{x}), or molybdenum silicon oxide (MoO\textsubscript{x}Si\textsubscript{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 SiO\textsubscript{2}. It is suspected that the Si 2p peak at ~102 eV is due to the MoO\textsubscript{x}Si\textsubscript{y} bonding\textsuperscript{45}. The Si 2p binding energy at ~102 eV is very close to that of (MoO\textsubscript{3})\textsubscript{70}(SiO\textsubscript{30}) (102.5 eV)\textsuperscript{45}. We later discuss that the interfacial layer can alter the electrical properties of MoS\textsubscript{2} film. The XPS depth profiling was also performed for a very thick MoS\textsubscript{2} film (Figure S8) and observed results are also similar to few-layer MoS\textsubscript{2}.

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 ‘SiO\textsubscript{2}’ are detected at ~99 eV and ~105 eV, respectively, and ‘O’ is detected at ~525 eV. Therefore these two points are clearly SiO\textsubscript{2}. 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 MoS\textsubscript{2}. 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 MoS\textsubscript{2}/SiO\textsubscript{2}. It is suspected that during sputtering process, Mo adlayers are initially formed at the interface of MoS\textsubscript{2}/SiO\textsubscript{2} and the Mo layers diffused into SiO\textsubscript{2} during
the annealing step, resulting in the formation of MoOxSi 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 increases46,47. 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 literature48,49. 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 shift52,53 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 MoS218,36 (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 (Ra, 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 (Ra) 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
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 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⁺ 3d⁵/₂ and 3d³/₂ 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.
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 MoS$_2$ films with ~1 $\times$ 9 cm$^2$ area and its Raman spectra are shown in Figure S13.

We have fabricated MoS$_2$ FETs and performed I–V measurement to investigate electrical properties. The schematic diagram of MoS$_2$ 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 MoS$_2$ film exhibited very high resistance in the range between 16 G$\Omega$ and 0.2 G$\Omega$. $I_d$–$V_g$ and $I_d$–$V_d$ plots of these devices are presented in Figure S14a–f. Our previous results showed that as-sputtered MoS$_2$ at RT are amorphous structure and are oxidized. As a result, as-sputtered film can exhibit 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 MoS$_2$ FET (1 min-sample). The field-effect mobility was extracted based on the slope of $\frac{\Delta I_d}{\Delta V_g}$ fitted to the linear regime 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 cm$^2$/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 MoS$_2$ 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 $\sim$6 times greater than bilayer MoS$_2$ (1 min-sample). The current on/off ratio values are $\sim$2 $\times$ 10$^3$ –4 $\times$ 10$^4$ for few-layer MoS$_2$ FETs. The extracted field-effect mobility is $\sim$181 and $\sim$173 cm$^2$/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 MoS$_2$ FETs. A significant enhancement can be noted in our MoS$_2$ FETs. It is interesting to compare with the recent reported mobility of $\sim$12 cm$^2$/Vs for thin MoS$_2$ film, but the mobility decreased significantly to $\sim$0.44 cm$^2$/Vs for
~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 MoS₂, and ~17 cm²/Vs for CVD-grown single crystal bilayer MoS₂. 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. 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 materials.

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.
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 MoOxSiy 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-MoS2.

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 MoOxSi thin sheets 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 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 compared the hysteresis under vacuum environment to prevent such extrinsic and environmental effects and focus on the trapping at the MoS$_2$/SiO$_2$ interface$^{60}$. The exfoliated-MoS$_2$ and CVD-grown MoS$_2$ exhibited large hysteresis in there Id–Vg curves. On the contrary, the sputtered-MoS$_2$ film exhibited small hysteresis. Such improvement in the hysteresis can be attributed to the small trap at the MoS$_2$/SiO$_2$ interface of the sputtered-MoS$_2$ 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-MoS$_2$ film.

Conclusions
We have successfully demonstrated the growth of large-area and continuous bilayer to few-layer MoS$_2$ on SiO$_2$/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 MoS$_2$ FETs exhibited a high field-effect mobility of $\approx$29 cm$^2$/V·s and an on/off ratio of $\approx$$10^4$. The mobility value of our bilayer MoS$_2$ FETs is larger than any of latest results of single to bilayer MoS$_2$ grown on a SiO$_2$/Si substrate with a SiO$_2$ gate oxide. The mobility for few-layer MoS$_2$ FETs increased to $\approx$173–181 cm$^2$/V·s. Our few-layer MoS$_2$ FETs exhibited the highest mobility value ever for any MoS$_2$ FETs with a SiO$_2$ 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$_x$Si$_y$ layer. The combined synthesis route of MoS$_2$-RF sputtering with the post-deposition annealing process could open up the possibility of mass and batch production of MoS$_2$ film. We believe our proposed strategy will pave the way for applications of MoS$_2$ in future electronics and optoelectronics.

Method
The various sizes of SiO$_2$ (300 nm)/Si substrates ranging from $1 \times 1$ cm$^2$ to $3 \times 3$ cm$^2$ 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 SiO$_2$/Si substrates into a sputtering chamber, the chamber was vacuumed at $1 \times 10^{-6}$ Torr. Before the deposition process, the MoS$_2$ 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 MoS$_2$ 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 MoS$_2$ 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

Figure 8. (a) Id–Vd of MoS$_2$ FET of 1-min sample; (b) Id–Vg of MoS$_2$ FET (1 min-sample) at Vd = 1, 3, 4 and 5 V; (c,d) Id–Vg of MoS$_2$ FETs, 3 min-sample (c), and 5 min-sample (d) at fixed Vd = 1V.
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.

Characterization details of MoS₂ films. Synthesized MoS₂ films were analyzed by Raman spectroscopy (Renishaw invia RE04, 512 nm Ar laser) with a spot size of around 1 μm and 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 Kα, 6.7 × 10⁻⁸ Pa) and photoluminescence (PL) with a 512 nm wavelength was used. Laser radiation of PL was focused onto the MoS₂ film with a spot-size of around 1 μm, (FFT, IFFT, etc.) were performed using the Gatan Digital Micrograph software (Gatan Microscopy Suite 2.0).

Fabrication of the MoS₂ FET devices. The active area of MoS₂ 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 MoS₂ transistors were measured using the 2 probe method at room temperature in a vacuum chamber to avoid oxidation.

| S. No. | Growth method | Lᵣ/Lᵢ(off) | Mobility [cm²/Vs] | Ref. |
|--------|---------------|------------|------------------|-----|
| 1      | Sputtering (MoS₂) + CVD | ~10⁴ | ~29 (~1.4 mm) – 173 ~ 181 (~3.8 ~ 6 mm) | This work |
| 2      | CVD (MoO₃ + S) | ~10⁷ | 24 | Appl. Phys. Lett., 106 (2015) 062101 |
| 3      | Sputtering (MoS₂) | 12.2 | 0.003–0.03 | Nanoscale, 2015, 2497~2503 |
| 4      | MoO₃ powder + Mo substrate+ CVD | 192 | 2–7 | Appl. Phys. Lett., 105 (2014) 072105 |
| 5      | Ebeam (Mo) + CVD | 12.2| 0.004–0.04 | ACS Appl. Mater. Interfaces, 2014, 6 (23), 21215–21222 |
| 6      | Sputtering (Mo)+ CVD | ~1.5 × 10⁴ –5 × 10⁴ | 0.01–0.7 | J. Amer. Chem. Society 2013, 135, 5304. |
| 7      | CVD (MoCl₃ + S) | ~10⁻⁷–10⁻⁵ | 0.09 | Nano Research 2014, 7 (12) : 1759–1768 |
| 8      | CVD (MoO₃ + S) on rGO | ~10⁴ | 0.02 | Adv. Mater. 2012, 24, 2320–2325 |
| 9      | CVD (MoO₃ + S) | ~10⁸ | 2–7 | ACS Nano, 2014, 8 (6), 6024–6030 |
| 10     | Ebeam (Mo) + CVD | 12 | 0.004–0.04 | Small 2012, 8, 966. |
| 11     | CVD (MoO₃ + S) | ~10⁸ | 17 | Appl. Phys. Lett. 100, 123104 (2012) |
| 12     | CVD (MoO₃ + S) | ~10⁻⁷–10⁻⁶ | 0.1–0.7 | ACS Nano, 2014, 8 (6), 6024–6030 |
| 13     | CVD (MoO₃ + S) | ~10⁴ | 0.09 | Nano Research 2014, 7 (12) : 1759–1768 |
| 14     | Thermal (MoO3) + CVD on sapphire | ~10⁴ | –0.8 | Nanoscale, 2012, 4, 6637–6641 |
| 15     | Thermolysis of (NH₄)₂MoS₄ | ~10⁷ | 4.7–6 | Nano Lett., 2012, 12 (3), 1538–1544 |
| 16     | Exfoliated (electrochemical) | ~10⁴ | 1.2 | ACS Nano, 2014, 8 (7), 6992–6910 |
| 17     | CVD (H₂S + Mo) | ~10⁸ | 0.12 | Nanoscale, 2014, 6, 2821–2826 |
| 18     | Mo(CO)₆ + (C₂H₅)₂S | ~10⁴ | 30 | Nature, 2015, 520, 656–660 |
| 19     | CVD (MoO₃ + S) | ~10⁻⁶ | 3.6 (1L), 8.2 (2L), 15.6 (3L) | ACS Nano, 2015, 7, 1688–1695 |
| 20     | CVD(MoO₃ + S) | ~10⁻⁷–10⁻⁴ | 3 to 4 | Nat. Mater., 2013, 12, 554–561. |

Table 1. Literature values of room temperature field-effect mobility for MoS₂ 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 MoS₂ FET devices. The active area of MoS₂ 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 MoS₂ transistors were measured using the 2 probe method at room temperature in a vacuum chamber to avoid oxidation.

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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. I.E., W.-G.L. and J.J. Participation included planning, experimental work and discussion. All authors read and approved the final manuscript.

**Additional Information**

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