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Sulfurization of as-deposited epitaxial MoO$_2$ films with high density of defects results in textured MoS$_2$ films. A capping layer annealing process is developed to improve the MoO$_2$ precursor film quality, resulting in lower defect density and better surface roughness. Sulfurization of the improved MoO$_2$ precursor film results in single-crystalline MoS$_2$ film instead of textured films, with much better device performance.
Enhanced Quality of Wafer-Scale MoS 2 Films by a Capping Layer Annealing Process

Xiangming Xu, Chenhui Zhang, Mrinal K. Hota, Zhixiong Liu, Xixiang Zhang, and Husam N. Alshareef* 

Wafer-scale, single-crystalline 2D semiconductors without grain boundaries and defects are needed for developing reliable next-generation of integrated 2D electronics. Unfortunately, few literature reports exist on the growth of 2D semiconductors with single-crystalline structure at the wafer scale. It is shown that direct sulfurization of as-deposited epitaxial MoO 2 films (especially, with thickness more than ~5 nm) produces textured MoS 2 films. This texture is inherited from the high density of defects present in the as-prepared epitaxial MoO 2 film. In order to eliminate the texture of the converted MoS 2 films, a new capping layer annealing process (CLAP) is introduced to improve the crystalline quality of as-deposited MoO 2 film and minimize its defects. It is demonstrated that sulfurization of the CLAP-treated MoO 2 films leads to the formation of single-crystalline MoS 2 films, instead of textured films. It is shown that the single-crystalline MoS 2 films exhibit field-effect mobility of 6.3 cm 2 V -1 s -1 , which is 15 times higher than that of textured MoS 2. These results can be attributed to the smaller concentration of defects in the single-crystalline films.

1. Introduction

The 2D MoS 2 is one of the most promising semiconductors for next generation electronics due to its tunable band gap, favorable electronic mobility, excellent optoelectronic properties, and good thermal and chemical and mechanical stabilities. Scalable fabrication of 2D films is the precondition for manufacturing of post-Moore’s law large-scale integrated devices. Researchers have already realized wafer-scale growth of MoS 2 films through chemical vapor deposition and metal organic chemical vapor deposition and vapor-solid reaction process. However, up to date, almost all the above growth processes could only realize polycrystalline MoS 2 films. Thus, most of these reported wafer-scale 2D MoS 2 films showed inferior electronic properties compared to single-crystalline MoS 2 films. In order to bring wafer-scale 2D MoS 2 electronic properties close to theoretical levels, it is necessary to explore new approaches to obtain single-crystalline structures at the wafer scale.

It has been reported that the structure and properties of wafer-scale MoS 2 films, grown by the two-step process, can be engineered by modifying the structure of the precursor film. For example, an epitaxial MoO 2 film resulted in quasi-single-crystalline MoS 2 films after sulfurization. However, still small angle grain boundaries and texture were observed in these MoS 2 films through direct sulfurization of the as-grown epitaxial precursor film. Here we show that thicker (>5 nm) precursor MoO 2 films lead to heavier texture in the converted MoS 2 films. The reason for using MoO 2 as a precursor film is because of its ability to grow continuous ultrathin film epitaxially on high-temperature stable substrates. In order to eliminate texture from the MoS 2 films and achieve single-crystalline MoS 2, we developed a new process to improve the crystalline quality of the epitaxial precursor MoO 2 film. This annealing process is henceforth referred to as the capping layer annealing process (CLAP). Through the sulfurization of the CLAP-treated epitaxial MoO 2 film, wafer-scale single-crystalline MoS 2 films were obtained without texture. Transistor performance showed that MoS 2 film with single-crystalline structure exhibits field effect mobility almost 15 times higher than that of the textured MoS 2.

2. Results and Discussions

2.1. MoS 2 Converted from CLAP-Treated Epitaxial MoO 2 Films

Texture in MoS 2 films derived from epitaxial MoO 2 precursor films is caused by a large number of defects such as misfit dislocations present in the as-grown epitaxial precursor MoO 2 film. Direct thermal annealing process has normally been used to reduce the defects and improve the crystalline quality of epitaxial precursor films. However, this approach usually produces noncontinuous films or films with rough surfaces.
due to recrystallization,[34–36] which we also observe upon direct-annealing our epitaxial MoO$_2$ films in this report. Thus, direct thermal annealing of the precursor epitaxial MoO$_2$ film is not a good approach for growing high-quality 2D MoS$_2$ films. To improve crystalline quality of the ultrathin epitaxial MoO$_2$ precursor film without losing the continuity and surface flatness, we have developed a new CLAP. This process has the capability of improving the quality of epitaxial precursor films without degrading their flat surface.[37]

In order to clarify the preparation process of wafer-scale single-crystalline MoS$_2$ film, a process flow diagram is shown in Figure 1a. First, a 2° (001) sapphire wafer was cleaned and placed in a pulsed laser deposition (PLD) chamber for MoO$_2$ deposition. The epitaxial film was realized with optimized PLD parameters, with film thickness of more than 4.5 nm, as shown in Figure 1b. Besides, the thickness could be controlled by adjusting the number of laser shots. After PLD, but before annealing, a transparent capping layer of Si$_3$N$_4$ (50 nm thick) was deposited on the MoO$_2$ film uniformly by plasma enhanced chemical vapor deposition. Then the Si$_3$N$_4$-protected MoO$_2$ sample was annealed at 900 °C for 1 h in Ar atmosphere. This CLAP-treated MoO$_2$ film is shown in Figure 1c. After CLAP, the sample was cooled down to room temperature naturally. After cooling, a buffered oxide etch (BOE) (20:1) solution was used to etch the Si$_3$N$_4$ capping layer. After removing the capping layer, a higher quality epitaxial MoO$_2$ film was obtained, with outstanding uniformity and continuity as shown in Figure 1d. Then, the MoS$_2$ sample was obtained after the sulfurization, as shown in the optical picture in Figure 1e. The above describes the process for the single-crystalline film preparation process. In contrast, only a textured MoS$_2$ film (Instead of a single-crystalline structure) could be obtained through direct sulfurizing...
of the as-deposited epitaxial MoO$_2$ film without CLAP. Detailed analysis of these two types of films is shown next.

2.2. Comparison of Directly-Annealed and CLAP-Treated Epitaxial MoO$_2$ Films

Epitaxial MoO$_2$ film was obtained by PLD using MoO$_3$ as target, 10 mTorr O$_2$ atmosphere, and 400 °C substrate temperature. Initially, we attempted to reduce the defects of the as-deposited MoO$_2$ film by direct thermal annealing (without capping layer) at different temperatures in Argon atmosphere. The sharpening of the X-ray diffraction (XRD) pattern and Raman peaks of the directly-annealed samples indicated an improved crystalline quality, as depicted in Figures S1 and S2 in the Supporting Information. But this direct annealing process degraded surface roughness and/or induced discontinuity in the MoO$_2$ films (Figure S3, Supporting Information), which is not acceptable for wafer-scale 2D film conversion. Thus, we developed the CLAP by introducing a protective layer to prevent film surface morphology from deteriorating. The protective layers used in CLAP, such as SiO$_2$ or Si$_3$N$_4$, are designed to be high-temperature stable and removable by BOE, a chemical that does not etch the MoO$_2$ film (Figure S4, Supporting Information). However, using SiO$_2$ capping layer did not perform as well as Si$_3$N$_4$ because some residual SiO$_2$ always remained on the MoO$_2$ film surface. The Si$_3$N$_4$ layer was easier to be totally removed, as indicated by the atomic force microscopy (AFM) phase images in Figure S5 in the Supporting Information. In addition, the Si$_3$N$_4$ capping layer resulted in smoother MoO$_2$ film surface, as depicted in Figure S6 in the Supporting Information.

The impact of the annealing process on the crystalline structure of epitaxial MoO$_2$ film was studied by XRD and Raman spectroscopy, as demonstrated in Figure 2a–c. Note that the crystal orientation of MoO$_2$ was not changed after the thermal treatment (with or without CLAP), as depicted in Figure 2a. The Rocking curve results are shown in Figure 2b. The full width at half maximum (FWHM) of the (200) peak was reduced from 0.085 to 0.072 as a result of the CLAP, meaning that the crystal quality of the epitaxial film was improved because of the thermal treatment. The slight peak shift to lower angle might be due to strain released during the annealing process. Figure 2c shows the Raman spectra of pristine, directly-annealed (uncapped) and CLAP-annealed MoO$_2$ films. The sharper peaks of both annealed MoO$_2$ films confirm their improved crystal quality. However, the Raman peak intensity of the CLAP-treated film at position 202 nm is stronger than that of directly-annealed (uncapped) film (position shift to 204 nm). In contrast, the peak intensity of CLAP-treated film at position 491.5 nm is much weaker (the peak position of the directly-annealed film shift to 498.5 nm). The reason might be that the vibration mode of the 202 peak is parallel to the (200) MoO$_2$ lattice plane, but the 491.5 peak is not in the same lattice plane. Since the CLAP resulted in continuous MoO$_2$ films (Figure 2f), an enhanced vibration mode was observed in the (200) lattice plane.$^{[38]}$ The Raman peak position shifts shown in Figure 2c (from 202 and 491.5 for CLAP film to 204 and 498.5 for directly-annealed uncapped film) might be due to two reasons: one is the different degree of phonon confinement in between the ultrathin CLAP-treated continuous film and the direct-annealed isolated MoO$_2$ grains$^{[39]}$; the other reason is the different amount of strain$^{[40]}$ released from these two films. Figure 2d–f shows the AFM images of pristine (as-deposited), directly-annealed (uncapped), and CLAP-treated epitaxial MoO$_2$ film, respectively. It is apparent from Figure 2e that film annealing with no capping layer results in discontinuous micrograins. The film treated by the CLAP in Figure 2f remains continuous and flat (room mean square (RMS) roughness = 0.342 nm). This value slightly increases compared to 0.167 nm of the pristine film (Figure 2d). This roughness might result from the plasma surface treatment during the capping layer deposition$^{[41]}$ or from enhanced atomic movement during the high-temperature annealing process.$^{[42]}$ Figure 2g shows a schematic of the MoO$_2$ film morphology variation with increasing direct annealing temperature. The as-prepared MoO$_2$ film is full of misfit dislocations or mosaic disorientation defects, as indicated in the enlarged lattice structure figure I: The MoO$_2$ film initially becomes rough at lower annealing temperature. II: Further increasing the temperature induces MoO$_2$ film recrystallization and forms interconnected micrograins. III: When the direct annealing temperature reaches 900 °C, the micrograins become isolated. Figure 2h shows a schematic of the different stages of the CLAP. Non-differently, the as-deposited MoO$_2$ film has a higher density of defects such as dislocation and mosaicity. Before the thermal annealing process, a capping layer (e.g., Si$_3$N$_4$) is deposited on top of the MoO$_2$ film. A thermal annealing process is then carried out which sharply annihilates defects and improves the epitaxial quality of the MoO$_2$ film. Meanwhile, mass transfer on the film surface caused by recrystallization is effectively prevented by the capping layer, leading to smoother and continuous films.

2.3. Microscopic Characterization of MoS$_2$ Films

In order to analyze the influence of the quality of the epitaxial precursor MoO$_2$ films on the quality of the converted MoS$_2$ films, the conversion process was carried out at fixed conditions. The conversion process detail is provided in the Experimental Section. The transmission electron microscopy (TEM) and scanning TEM (STEM) images shown in Figure 3 were all obtained from MoS$_2$ films with thickness of ∼5 nm. Low magnitude TEM image characterization was performed on the MoS$_2$ film converted from the CLAP-treated MoO$_2$ film; the three areas marked by red circles (Figure 3a) show selected area electron diffraction (SAEDs). The dark area indicated by the yellow arrow is the Cu grid, and the gray network indicated by the red arrow is the lacy carbon region. Figure 3b shows the low magnification STEM image of the MoS$_2$ film converted from CLAP-treated MoO$_2$ film. From Figure 3b we could find that the whole
continuous MoS₂ film has only one or two discontinuous monolayers on the top surface. Figure 3c shows a high magnification STEM image focused on the edge area of the top discontinuous monolayers. The inset fast Fourier transform (FFT) pattern shows a single-crystal like pattern, indicating that the crystalline orientation of the top discontinuous monolayers is consistent
with that of underneath continuous film. The morphology of this kind of film is further illustrated with side and top views in Figure 3e,f, respectively. The HR STEM high-angle annular dark-field (HAADF) image (Figure 3d) of the stable single-crystalline MoS$_2$ film depicts the honeycomb structure with AA' stacking order of the 2$H$ MoS$_2$ phase.$^{[44]}$ The comparison between AA' and AB staking mode is shown with side and top views in Figure 3g. Figure 3h–j displays the SEADs taken at three locations marked (1, 2, and 3) in Figure 3a. All diffraction patterns exhibit hexagonally arranged diffraction spots, demonstrating the single-crystalline structure of the MoS$_2$ films. Since these three selected areas were all set within a diameter of about 20 µm (the upper limit of our TEM instrument), they illustrate that the single-crystalline structure uniformity is exhibited at the wafer scale. Figure 3k shows the SAED pattern (location marked with red circle in Figure S7 in the Supporting Information) of the MoS$_2$ converted from pristine precursor epitaxial MoO$_2$ film without any postannealing treatment. The ring-like pattern consists of quasi discrete elongated light spots, indicating the preferred orientation and the texture structure in the 2D MoS$_2$ film. Several researchers have demonstrated that these kind of structures have great influence on the mechanical, optical, and electronic properties of thin films.$^{[45–48]}$ However, this is probably the first time that textured morphology has been observed in 2D chalcogenide films. We believe that the reason for forming the texture in 2D films is the presence of high density of defects in the precursor MoO$_2$ films. Achievement of the single-crystalline structure can be attributed to the high-quality precursor film obtained after the CLAP treatment. TEM characterization of thicker (15 nm) MoS$_2$ films converted from pristine and CLAP-treated precursor were also performed, as depicted in Figure S8 in the Supporting Information. The SAED (Figure S8b, Supporting Information) of the 15 nm thick film from pristine precursor displayed textured structure, with diffraction pattern that resembles the ring pattern of textured films shown in Figure 3k. However, the thicker MoS$_2$ films converted from the CLAP-treated precursor retain a perfect single-crystalline structure (Figure S8d, Supporting Information).

**2.4. Spectral Characterization**

The Raman, photoluminescence (PL), and X-ray photoelectron spectroscopy (XPS) measurements were performed on the...
MoS$_2$ films converted from the CLAP-treated or pristine MoO$_2$ films. The intensity-normalized Raman spectra in Figure 4a show that no peak shift between textured and single-crystalline MoS$_2$. Further, no peak shift is observed among single-crystalline films with different thickness. The peak differences between $E_{2g}$ and $A_{1g}$ peaks all are 25.4 cm$^{-1}$, which means that all the films mentioned above show bulk film structure (film with more than five layers or 3 nm) and no strain difference. A total of 33 locations on the 4.5 nm thick MoS$_2$ film wafer (Figure S9a, Supporting Information) were tested by Raman. All tested locations show the same $E_{2g}$ and $A_{1g}$ peak positions (Figure S9b, Supporting Information). This result confirms the film uniformity across the 2″ wafer. In Figure 4b, The PL spectra of 15 nm MoS$_2$ films reveal that the FWHM of PL peak at 676 nm position of single-crystalline film is smaller than that textured films, which means single-crystalline MoS$_2$ films exhibit better quality. Typical XPS spectra of textured MoS$_2$ with thickness 4.5, 6, and 9 nm are shown in Figure 4c–d. Figure 4c shows XPS peaks at 235.88, 232.73, 232.83, 229.67, and 226.87 corresponding to Mo$^{6+}$ 3d$_{3/2}$, Mo$^{6+}$ 3d$_{5/2}$, Mo$^{4+}$ 3d$_{3/2}$, Mo$^{4+}$ 3d$_{5/2}$, and S 2s, respectively. Figure 4d shows S 2p$_{1/2}$ and S 2p$_{3/2}$ peaks at 163.70 and 162.51 eV. These peak positions are consistent with those reported for crystalline MoS$_2$ films.[21] All the single-crystalline and textured MoS$_2$ films with different thickness actually show the same XPS peak positions as illustrated above. Through careful Lorentzian–Gaussian fitting of XPS, The Mo/S elemental ratio in films with thickness 4.5, 6, and 9 nm were calculated to be 1/1.92, 1/1.91, and 1/1.89, respectively. This data indicates that the thicker precursor MoO$_2$ films are more difficult to sulfurize, hence the resultant MoS$_2$ films contained more defects. This effect originate from the difficulty of sulfur elements replacing oxygen (defined as S-O-exchange defects) deeper in the thick MoO$_2$ films.

2.5. Transfer Process and Transistor Performance

The MoS$_2$ films deposited with different thicknesses on 2″ wafers could be uniformly transferred onto both rigid and flexible substrates using PDMS stamp transfer combined with wet chemical etching process. The image in Figure 5a shows transferred films with thickness 4.5 and 6 nm (as shown by the inset AFM images and line scan) on SiO$_2$(300 nm)/Si$^+$ substrates. The films were also successfully transferred onto flexible paper substrate as displayed in Figure 5b. The transfer process above demonstrates the potential for our wafer-scale MoS$_2$ films to be used in Si-based, flexible, and paper electronics.

Figure 4. a) Raman, b) PL, c) XPS-Mo 3d, d) XPS-S 2p spectra of single-crystalline and textured MoS$_2$ films converted from CLAP-treated and pristine MoO$_2$ films with different thickness.
Top-gated thin film transistors (TFTs) were fabricated, as shown in Figure 5c, to characterize the electronic properties of the MoS$_2$. The S/D (source and drain) and top gate Au/Ti (50/10 nm) contacts were deposited by e-beam evaporation at room temperature in high vacuum. HfO$_2$ gate dielectric (60 nm thick) was deposited using atomic layer deposition (ALD). Details of the TFT fabrication are explained in the Experimental section. The output curves of TFTs fabricated using single-crystalline and textured MoS$_2$ films (both 6 nm thick) are shown in Figure 5d, where Ohmic-like behavior with S/D electrodes could be observed. Transfer curves of TFTs fabricated using both single-crystalline MoS$_2$ (red curves) and textured MoS$_2$ (black curves) channel with different thicknesses (4.5, 6, and 9 nm) are plotted along with conductivity on a logarithmic scale (Figure 5e). All the transfer curves were measured under the same S/D voltage (1 V) for a fair comparison. The results shown in Figure 5f (also Figure S10a,b in the Supporting Information) indicate that the single-crystalline MoS$_2$ based TFTs always show higher mobility than textured MoS$_2$ based TFTs, no matter the channel thickness. Especially, when the MoS$_2$ film thickness was 4.5 nm, the single-crystalline MoS$_2$ film showed a mobility of 6.3 cm$^2$ V$^{-1}$ s$^{-1}$ (this result is better than that of the MoS$_2$ films converted from the other Mo-based precursors),$^{[22–24,49]}$ which is 15 times higher than that of the textured MoS$_2$. This result is likely due to the high concentration of defects in the textured films which can scatter the channel electrons. Subthreshold swing values (Figure S10c, Supporting Information) did not show any clear trend, probably because of fluctuations in the TFT fabrication process. As the films become thicker, the TFT performance with both films (single-crystalline MoS$_2$ and textured MoS$_2$) show significant degradation, and the mobility differences between the single-crystalline and textured MoS$_2$ become smaller. The degraded mobilities and current on/off ratios measured for thicker MoS$_2$ film devices are in contrast to the trend normally observed in mechanically-exfoliated or CVD-grown flakes.$^{[50,51]}$ The reason for this difference might be that at larger MoS$_2$ film thickness both the single-crystalline and textured MoS$_2$ films have more S-O-exchange defects. These defects could act as electrons-scattering centers, but more experimental work is needed to confirm this hypothesis.

3. Conclusion

Single-crystalline MoS$_2$ films were grown on $2''$ wafers by sulfurization of capp-annealed MoO$_2$ films. Our CLAP could eliminate defects present in the as-deposited epitaxial MoO$_2$
precursor films, which resulted in single-crystalline MoS2 films. However, the direct sulfurization of as-deposited epitaxial MoO2 films (without capping layer) resulted in textured MoS2 films. The distinction between the single-crystalline and textured MoS2 films was clarified through TEM characterization. The wafer-scale MoS2 films of different thickness were successfully transferred onto rigid SiO2 (300 nm)/Si++ and flexible paper substrates. Electronic properties of the single-crystalline and textured MoS2 films were compared. Transistors fabricated single-crystalline films showed 15 times higher field effect mobility than their textured counterparts.

4. Experimental Section

Epitaxial MoO2 PLD Deposition: (001) Al2O3 substrate was precleaned and thermally annealed in vacuum for 2 h to improve surface quality. The MoO2 deposition process was carried out at 400 °C in O2 ambient at a pressure of 10 mTorr. Thickness of MoO2 was controlled by the number of laser shots and by laser energy. This process is similar to the previous report.[26] CLAP Process: Pristine (as deposited) epitaxial MoO2 sample was covered with protective capping layer, such as SiO2 or Si3N4 with thickness of 50 nm. The SiO2 layer was deposited at room temperature by sputtering process. Si3N4 layer was deposited at 250 °C by plasma-enhanced chemical vapor deposition. The protective capping layer was thermally stable without crystallization even at temperature up to 900 °C. The MoO2 sample with capping layer with annealed in the PLD chamber at 900 °C for 1 h in Argon at a pressure of 50 mTorr. The annealing time cannot be prolonged to more than 2 h, as this would cause serious film uniformity issues. After the annealing process, the sample was cooled down to room temperature, and then put into BOE (20:1) for 2 h to totally remove the capping layer. It is made sure the selected capping layer etchant did not damage the MoO2. The bare MoO2 samples were then cleaned in DI water three times. Finally, the high-quality epitaxial MoO2 films with fewer defects were ready for the sulfurization process. Sulfurization Process: Three zone chemical vapor deposition system is used to do the sulfurization. During the sulfurization process, Argon carrier gas with flow rate of 100 sccm and pressure 5–10 torr was used. Sulfur powder source with 500 mg was put in the up-stream heating zone with temperature 150 °C. The MoO2 sample was put in the down-stream heating zone. After the temperature of the sulfur source stabilized at 150 °C, MoO2 sample was heated at a heating rate of 20 °C min−1. The sulfurization temperature was carried out at 900 °C for 1 h. Once the process finished, the system was naturally cooled down.

Top-Gated TFT Device Fabrication: The MoS2 films were used as channel layer and patterned by dry etching. An Au/Ti film with thickness (50/10 nm) was deposited by electron beam evaporation and used as S/D contact and top gate electrodes, patterned by lift-off process. HfO2 high-k dielectric layer, with thickness of 60 nm was deposited using atomic layer deposition. The via hole was patterned by dry etching.

Characterization: 2θ–2θ scans and Rocking curves X-ray diffraction patterns of the epitaxial MoO2 films were obtained using X-Ray Diffractometer (Bruker, D8 φ 2000) with Cu Kα (λ = 1.5418 Å). Raman and PL measurement was realized with Horiba Aramis Raman spectrometer (Horiba Scientific) equipment, excited by visible light with wavelength of 473 nm, AFM (Bruker, Dimension Icon SPM) was for the surface morphologies characterization of pristine, thermally-treated epitaxial MoO2, and transferred MoS2 films. TEM images and selected area electron diffraction patterns were obtained by transmission electron microscope (Titan Cs Probe, FEI). HR TEM images were performed in a Cs-corrected FEI Titan Themis microscope. The free-standing MoS2 film was obtained through 2 h etching in aqueous alkaline solution, then was transferred onto TEM grids with lacey carbon film. XPS spectra were measured in a Kratos Axis Ultra DLD spectrometer equipped with a monochromatic Al Kα X-ray source (hv = 1486.6 eV) operating at 150 W. All spectra were recorded using an aperture slot of 300 μm × 700 μm, while high-resolution spectra were measured with a pass energy of 20 eV and a step size 0.1 eV. Performance of top-gated TFTs was measured using an Agilent B1500A analyzer.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

MoS2, single crystalline, texture, transistor, wafer scale

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