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Probing the Growth Improvement of Large-Size High Quality Monolayer MoS₂ by APCVD

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Abstract: Two-dimensional transition metal dichalcogenides (TMDs) have attracted attention from researchers in recent years. Monolayer molybdenum disulfide (MoS₂) is the direct band gap two-dimensional crystal with excellent physical and electrical properties. Monolayer MoS₂ can effectively compensate for the lack of band gap of graphene in the field of nano-electronic devices, which is widely used in catalysis, transistors, optoelectronic devices, and integrated circuits. Therefore, it is critical to obtain high-quality, large size monolayer MoS₂. The large-area uniform high-quality monolayer MoS₂ is successfully grown on an SiO₂/Si substrate with oxygen plasma treatment and graphene quantum dot solution by atmospheric pressure chemical vapor deposition (APCVD) in this paper. In addition, the effects of substrate processing conditions, such as oxygen plasma treatment time, power, and dosage of graphene quantum dot solution on growth quality and the area of the monolayer of MoS₂, are studied systematically, which would contribute to the preparation of large-area high-quality monolayer MoS₂. Analysis and characterization of monolayer MoS₂ are carried out by Optical Microscopy, AFM, XPS, Raman, and Photoluminescence Spectroscopy. The results show that monolayer MoS₂ is a large-area, uniform, and triangular with a side length of 200 μm, and it is very effective to treat the SiO₂/Si substrate by oxygen plasma and graphene quantum dot solution, which would help the fabrication of optoelectronic devices.

Keywords: monolayer MoS₂; APCVD; Raman spectra; growth condition; PL spectrum; graphene quantum dot; Oxygen plasma

1. Introduction

Molybdenum disulfide (MoS₂) has attracted attention from researchers in recent years because of its excellent physical properties. MoS₂ has been studied extensively in the field of two-dimensional nano-electronic devices, which has the potential to become an alternative to graphene [1–4]. MoS₂ is a two-dimensional transition metal dichalcogenides (TMDs) with an adjustable band gap structure in which atoms in layers are bonded by covalent bond interaction, whereas interlayer atoms are coupled by weak van der Waals forces [5,6]. Monolayer MoS₂ has excellent photoelectric properties due to its high electron mobility and unique band gap structure. The bulk MoS₂ is an indirect bandgap semiconductor with band gap energy about 1.29 eV, band gap energy increases with the decrease of MoS₂ layers number, thus monolayer MoS₂ becomes the direct bandgap semiconductor with 1.90 eV [7–9]. Due to the existence of a direct band gap, field effect transistors based on monolayer MoS₂ have higher current switching ratio and electron mobility at room temperature [10,11]. In

addition, it can also be used in phototransistors [12], logic circuits [13], secondary batteries, and photo catalysis [14].

At present, most reports on the preparation of monolayer MoS₂ are by mechanical peeling [15], lithium ion intercalation [16–18], hydrothermal [19], and chemical vapor deposition (CVD) [3,4]. The above four methods are simple in operation, wide in range, but require high preparation of the environment, high cost, and have poor repeatability [20–22]. Moreover, the layer number of MoS₂ is difficult to control, and the maximum area of monolayer MoS₂ is 100 μm approximately according to existing reports, which is a disadvantage to the fabrication of optoelectronic devices, as shown in Table 1. Although the growth method of MoS₂ is already defined, the preparation of large-area uniform monolayer MoS₂ is still a difficult problem to be solved.

It can be seen from the above reported literature that the maximum side length of monolayer MoS₂ is 100 μm approximately [23,24]. Effects of the substrate processing conditions, such as oxygen plasma treatment time, power, and amount of graphene quantum dot solution on growth quality and area of MoS₂ are researched systematically in this paper. The large-area uniform triangle monolayer MoS₂ with a side length of 200 μm is prepared on an SiO₂/Si substrate by atmospheric pressure chemical vapor deposition (APCVD) without vacuum treatment, which can provide materials for the photovoltaic devices effectively [25]. First, this paper introduces the growth experiment of MoS₂ and testing equipment and then introduces the influence of different SiO₂/Si substrate treatment factors on growth morphology and quality of MoS₂. This is followed by the characterization of monolayer MoS₂ under optimal treatment conditions of SiO₂/Si substrate, which was carried out by Optical Microscopy, AFM, XPS, Raman, and Photoluminescence Spectroscopy. Finally, it is concluded that it is possible to obtain the large-area high-quality monolayer MoS₂ by treating SiO₂/Si substrate with oxygen plasma and graphene quantum dot solution, which would contribute to the preparation of photovoltaic device.

Table 1. Comparison of different growth modes of MoS₂.

Growth Method	Characteristics	Size of MoS ₂	References
Micromechanical stripping	Simple process, low yield, poor repeatability	10 μm	[15,16]
Lithium ion intercalation	Complicated operation and high cost	20 μm	[16–18]
Hydrothermal	Poor crystallization quality	20–30 μm	[19–21]
CVD	Layer number cannot control	30–50 μm	[2–4,8–10]
APCVD	Simple operation, no vacuum treatment	80–100 μm	[25]
APCVD	Oxygen plasma treatment, graphene quantum dot	200 μm	This paper

2. Methods

Before the MoS₂ growth experiment, SiO₂/Si substrates were respectively placed in absolute ethanol, acetone, and deionized water solution for ultrasonic cleaning in 10 min, 10 min, 10 min, and dried with a nitrogen gun. Compared to the reported literature, the difference is that SiO₂/Si substrates are cleaned with PCE-6 plasma cleaner under different time and power conditions. Subsequently, different dosages of 1 mg/mL graphene quantum dot solution were applied to SiO₂/Si substrates uniformly with oxygen plasma by using VTC-100 vacuum rotary coater. In addition, a quartz boat with 100 mg sulfur powder (Alfa Aesar, Shanghai, China, 99.5%) was transferred to the low temperature zone of the tube furnace constructed by the winding heating belt. Moreover, 2 mg MoO₃ powder (Alfa Aesar, Shanghai, China, 99.95%) was weighed into one end of the quartz boat, and SiO₂/Si substrate was placed faced down 5 cm apart from the MoO₃ powder, and the quartz boat with MoO₃ powder and SiO₂/Si substrate was placed in the high temperature zone of tube furnace, as shown in Figure 1a. Figure 1b shows the temperature change diagram of MoS₂ grown by APCVD, high-purity argon gas with a flow rate of 300 sccm was introduced into the tube furnace for 10 min to exclude air in the tube furnace before the start of the reaction. The high temperature zone of the tube furnace was heated from room temperature to 550 °C in 30 min, meanwhile the heating belt began to heat, and the temperature was controlled at 200 °C. The temperature of the high temperature zone continued to 720 °C in

10 min with heating, and maintained the growth temperature for 10 min before cooling down to room temperature. The temperature distribution in the furnace was as follows: The closer the distance from the intermediate heating zone of the tube furnace was, the higher the temperature was [26,27]. Finally, argon gas with a flow rate of 50 sccm was introduced into the tube furnace throughout the reaction experiment, and the pressure of the tube furnace was maintained at a normal pressure.

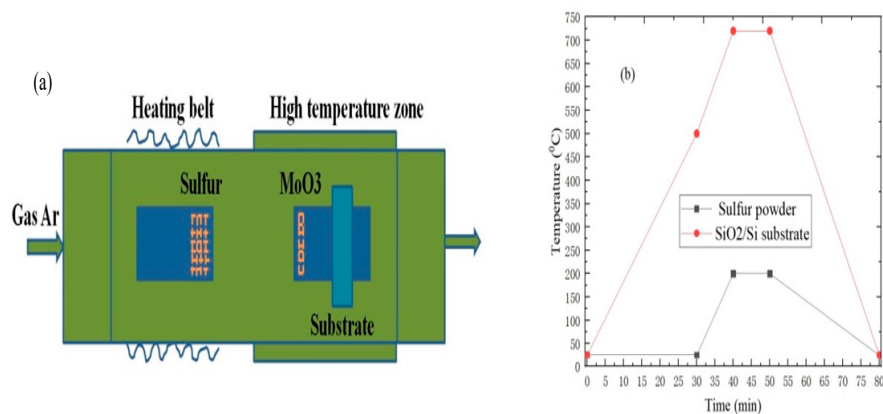


Figure 1. (a) Growth experiment schematic diagram of MoS₂; (b) temperature change diagram of MoS₂ grown by atmospheric pressure chemical vapor deposition (APCVD).

The new generation high-resolution Raman spectroscopy (LabRAM HR Evolution, HORIBA Jobin Yvon, Paris, France) was used to characterize the surface topography, Raman, and Photoluminescence spectrum of MoS₂ systematically. The specific test conditions of the Raman spectroscopy were as follows: 532 nm laser with 1 μm spot diameter, spectral resolution $\leq 0.65 \text{ cm}^{-1}$, scan time 5 s, and accumulations number 3 s. The layer number of MoS₂ could be determined quickly by the difference in the contrast between MoS₂ and SiO₂/Si substrate under the optical microscope. Raman spectrum was the scattering spectrum that utilized the inelastic scattering effect of the molecules on photons, which can provide an efficient representation of molecular structure. Photoluminescence spectrum is a form of cold luminescence, it would re-emit photons or electromagnetic waves when SiO₂/Si substance absorbs photons or electromagnetic waves of a certain frequency. In addition, surface morphology and thickness of MoS₂ grown on SiO₂/Si substrate were characterized through interatomic interaction forces by using atomic force microscopy with longitudinal high resolution (AFM, Dimension 3100, Veeco Instruments, Shanghai, China). Moreover, X-ray photoelectron spectroscopy-Auger electron spectroscopy (Theta 300 XPS system, Thermo Fisher, Shanghai, China) was also used to measure chemical and electronic states of MoS₂.

Optical microscopy (OM) was the most efficient and intuitive method to characterize layered structural materials. MoS₂ was grown on silicon substrate with a layer of 285 nm SiO₂ by APCVD. The number, size and film formation of MoS₂ can be quickly judged by the contrast between MoS₂ and SiO₂/Si substrate under the optical microscope.

3. Results and Discussion

3.1. Effect of Different Oxygen Plasma Treatment Time and Power on the Growth of MoS₂

SiO₂/Si substrates are cleaned by the PCE-6 oxygen plasma cleaning machine in this experiment. The specific cleaning steps are as follows: First put SiO₂/Si substrate into the vacuum chamber and vacuum it to below 1 Pa. Then oxygen gas is introduced to maintain oxygen vacuum in the vacuum chamber to 50 Pa. As the substrate treatment effect is greatly affected by gas pressure, this affects discharge power and processing time during glow discharge. Subsequently, the photo-discharge is performed at the discharge frequency of 13.56 MHz, and SiO₂/Si substrates are processed sequentially at 300 W, 500 W, and 1000 W discharge power for 30, 60, 90, and 120 s. The principle of oxygen

plasma cleaning is to convert oxygen into its plasma state by the glow discharge. These high-energy ions bombard the surface of the SiO_2/Si substrate and easily react with the surface of substrate to increase the surface energy, thereby changing the surface properties of the SiO_2/Si substrate and remove the organic impurities of the SiO_2/Si substrate surface, which can achieve a hydrophilic effect, and improve the surface activity of the silicon wafer [28]. The characterization of the SiO_2/Si substrate before and after oxygen plasma cleaning treatment can be observed by the growth of MoS_2 . The specific growth of MoS_2 under different oxygen plasma treatment time and power are as follows.

It can be found by observing Figure 2 that the growth and morphology of MoS_2 would, respectively, become uniform and better, and the size was also improved with an increase of processing time when the oxygen plasma was used in the low-power and medium-power oxygen plasma cleaning of SiO_2/Si substrate. This is because the surface roughness is improved significantly after oxygen plasma cleaning, and it can also change the surface characteristics of the SiO_2/Si substrate. The growth of MoS_2 initially becomes better and more uniform with the increase of processing time when the SiO_2/Si substrate is subjected to high-power oxygen plasma cleaning, but the treatment time has almost no effect on the growth of MoS_2 after 60 s, which indicates that too long processing time does not always increase the surface activity of the MoS_2 material. As the oxygen plasma cleaning power increases, the density and energy of the plasma increase, and the oxygen plasma treatment speed also increases. At this time, the morphology of MoS_2 is the triangle with uniform size, and multilayer or bulk materials of MoS_2 also appear when the time of cleaning SiO_2/Si substrate is constant. As shown in Figure 2, it is found that the optimal oxygen plasma cleaning power and time of SiO_2/Si substrate processing are 500 W and 90 s, respectively. At this time, the morphology of MoS_2 is the best, and the maximum triangular side length can be up to 100 μm .

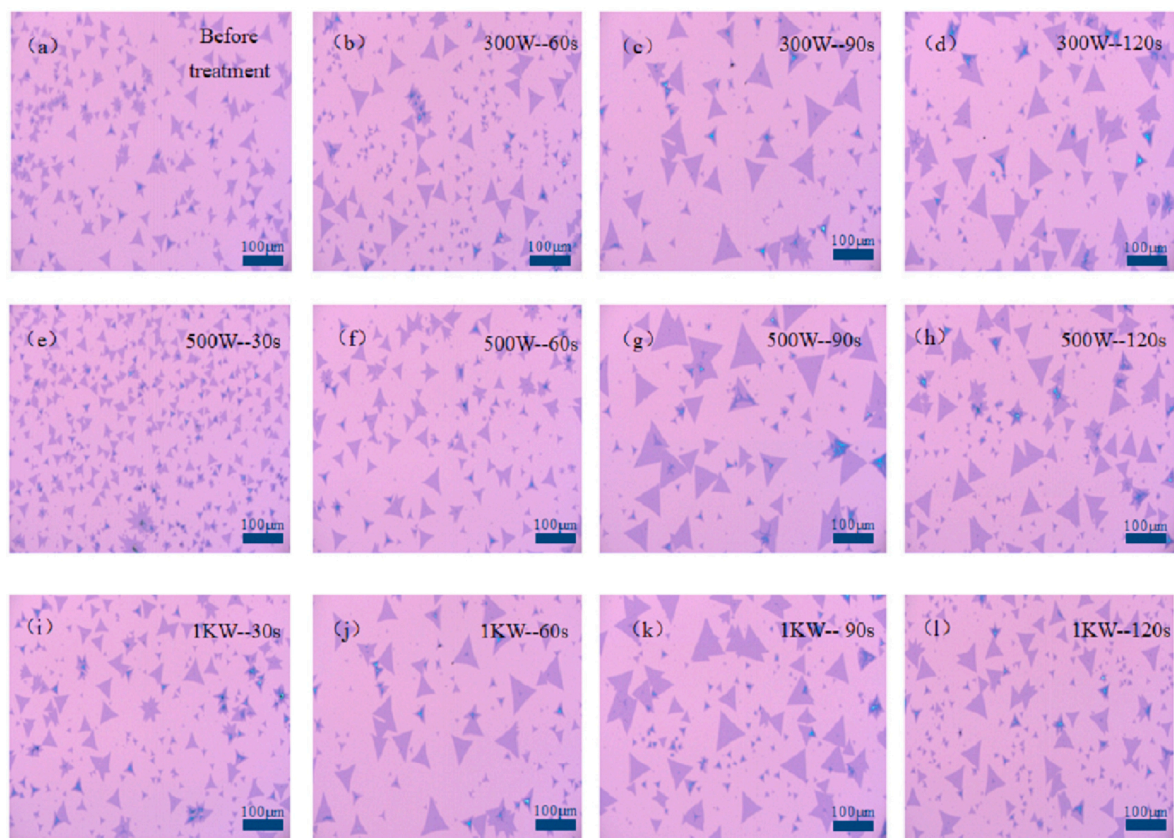


Figure 2. The growth of MoS_2 on SiO_2/Si substrate under the different oxygen plasma treatment time and power. (a) Before treatment, (b) 300 W, 60 s, (c) 300 W, 90 s, (d) 300 W, 120 s, (e) 500 W, 30 s, (f) 500 W, 60 s, (g) 500 W, 90 s, (h) 500 W, 120 s, (i) 1 KW, 30 s, (j) 1 KW, 60 s, (k) 1 KW, 90 s, (l) 1 KW, 120 s.

3.2. Effect of Graphene Quantum Dot Solution Amount on the Growth of MoS₂

Impurities and defects on the SiO₂/Si substrate are easily selected when MoS₂ sample is deposited by CVD. Therefore, the processing of SiO₂/Si substrate would have a greater impact on the nucleation process of MoS₂. VTC-100 vacuum rotary coating machine is used to spin the graphene quantum dot solution with a particle size of 2 nm on cleaned SiO₂/Si substrate uniformly in the experiment, and research the effect of different dosages of 1mg/ml graphene quantum dot solution on the growth of MoS₂ by non-uniform nucleation principle. The size, quality, and continuity of monolayer MoS₂ are improved by dropping the graphene quantum dot solution on SiO₂/Si substrate.

In order to obtain the optimum dosage of graphene quantum dot solution, SiO₂/Si substrates with 5 μ L, 10 μ L, 15 μ L, 20 μ L, 25 μ L, and 30 μ L of 1 mg/mL graphene quantum dot solution are carried out on the growth experiments of MoS₂. It can be seen from Figure 3 that different dosages of graphene quantum dot solution on SiO₂/Si substrates have an effect on growth of MoS₂. It can be observed from Figure 3a–d that many small triangles with a size of 20 μ m would appear on SiO₂/Si substrate under the same growth conditions, the size of the triangular MoS₂ increases gradually with graphene quantum dot solution dosage increase, large area continuous film with 300 μ m lateral dimension is formed, and the middle of the MoS₂ film is obviously thicker. This is because a certain dosage graphene quantum dot solution can provide a suitable nucleation point for MoS₂ on SiO₂/Si substrate, which would facilitate the growth of a large-area MoS₂ film. In Figure 3e,f, MoS₂ sample deposited on SiO₂/Si substrate is the triangle with a size of 40 μ m, and no uniform continuous film is formed. The reason is that the graphene quantum dots solution dosage is too high and the nucleation sites are excessive. In conclusion, the optimal dosage of 1 mg/mL graphene quantum dot solution is 20 μ L by analyzing the above results.

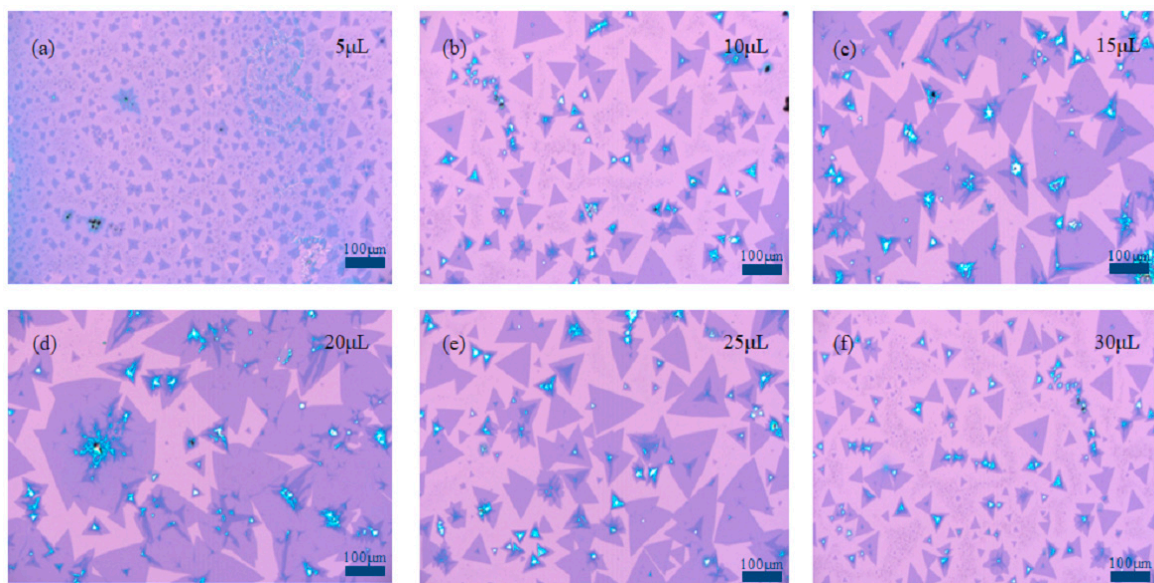


Figure 3. Effect of graphene quantum dot solution with different dosage on the growth of MoS₂. (a) 5 μ L, (b) 10 μ L, (c) 15 μ L, (d) 20 μ L, (e) 25 μ L, (f) 30 μ L.

3.3. The Optimal Growth Conditions of Monolayer MoS₂

In Figure 4, the size of large-area high-quality monolayer MoS₂ grown on SiO₂/Si substrate with oxygen plasma (500 w, 90 s) and graphene quantum dot solution (20 μ L, 1 mg/mL) can be up to 200 μ m, and different monolayer MoS₂ spliced together to form large area continuous film with uniform color and good film formation quality. The reason is that a certain dosage graphene quantum dot solution can provide a suitable nucleation point for MoS₂, which would contribute to the growth

of large-area monolayer MoS₂ film. Monolayer MoS₂ are characterized by Optical Microscopy, AFM, XPS, Raman, and Photoluminescence Spectroscopy. The following are the test results in detail.

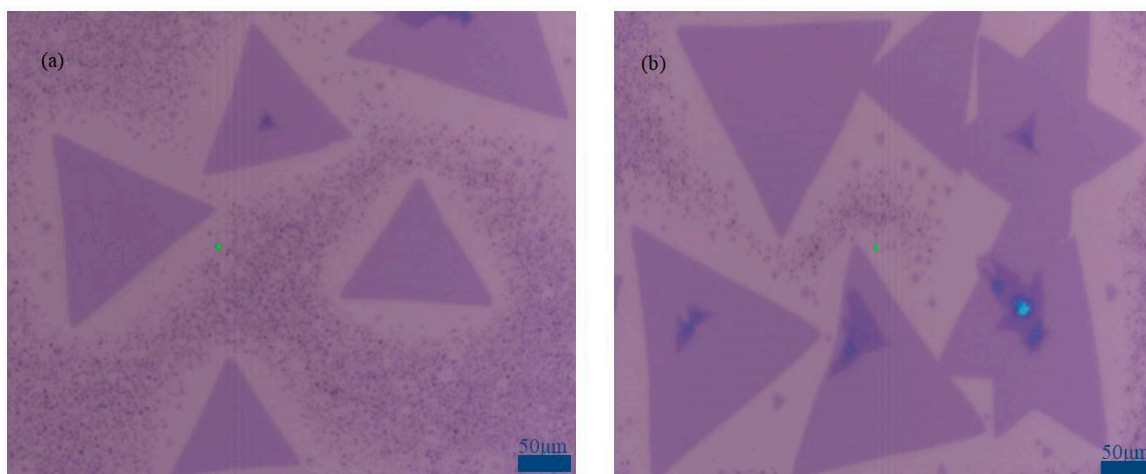


Figure 4. Growth of different regions MoS₂ on SiO₂/Si substrate under optimal treatment condition (a) first region; (b) second region Scale bar: 50 μm.

3.4. Characterization of Monolayer MoS₂

It can be found by observing Figure 5a that there are two characteristic peaks in Raman spectrum of monolayer MoS₂. The E_{2g}¹ peak position of in-plane vibration mode is located at 383.3 cm⁻¹, and the A_{1g} peak position of out-of-plane vibration mode is located at 402.7 cm⁻¹, the peak distance between two peaks is 19.4 cm⁻¹, and A_{1g}/E_{2g}¹ ≈ 1.05, which indicates the triangular MoS₂ is monolayer [29]. In Figure 5b, monolayer MoS₂ film has the strongest luminescence peak at 683.6 nm due to the direct excitation of excitons. It can be known from the conversion relationship between wavelength and electron volts that the corresponding transition energy level at the luminescence peak is about 1.82 eV, and there is also a B peak at 1.98 eV due to the interaction of electrons in the 3d orbital of molybdenum atoms, which can further prove that the sample is a large-area monolayer MoS₂ with good film quality [30,31]. In addition, Raman mapping is carried out to observe film-forming quality and uniformity of triangular monolayer MoS₂, and the color is very uniform, which also indicates that the sample is monolayer MoS₂ with highly uniform quality.

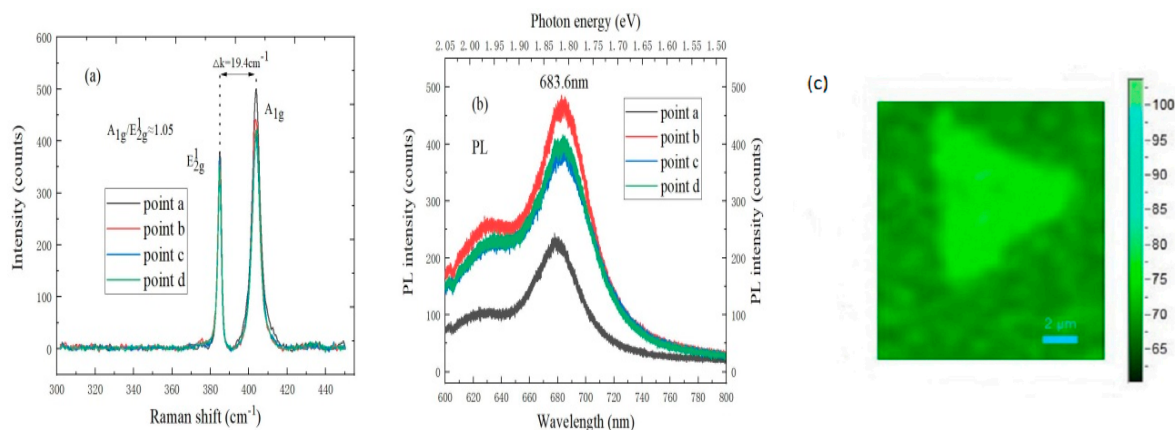


Figure 5. (a) Raman spectrum, (b) PL spectrum, and (c) mapping of monolayer MoS₂ under optimal treatment conditions of SiO₂/Si substrate.

Atomic force microscopy (AFM) is used to characterize the surface topography of layered structural materials. The thickness of MoS₂ on SiO₂/Si substrate is characterized by AFM, as shown in

Figure 6. The film and triangular MoS₂ are very uniform in color, and the thickness of MoS₂ is about 0.83 nm, which indicates that MoS₂ is monolayer.

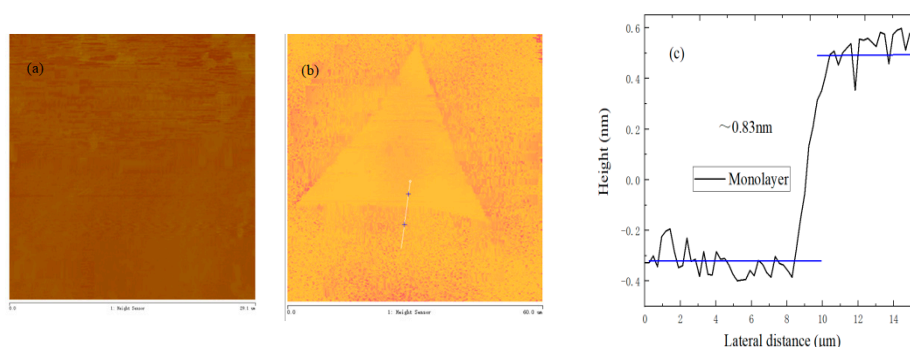


Figure 6. (a) Atomic force microscopy (AFM) topography of MoS₂ film; (b) AFM topography of triangular MoS₂; (c) AFM height profile of triangular MoS₂ (blue lines represent the average height of triangular MoS₂ under different position).

X-ray photoelectron spectroscopy (XPS) is the advanced analytical technique in microscopic analysis of electronic materials and components. It uses photoelectron binding energy as abscissa and relative intensity as ordinate to make photoelectron spectroscopy. The relevant information of MoS₂ is obtained by analyzing the energy spectrum. It can be seen from Figure 7 that Mo3d_{5/2} and 3d_{3/2} binding energy in Mo₄₊ is about 228.5 eV and 232.5 eV, respectively. At the same time, 2P_{3/2} and 2P_{1/2} binding energy in S_{2p} is about 162.2 eV and 163.3 eV, respectively. The percentages of the S_{2p} element and Mo_{3d} element are 15.79% and 7.58%, respectively, which also indicates MoS₂ is monolayer.

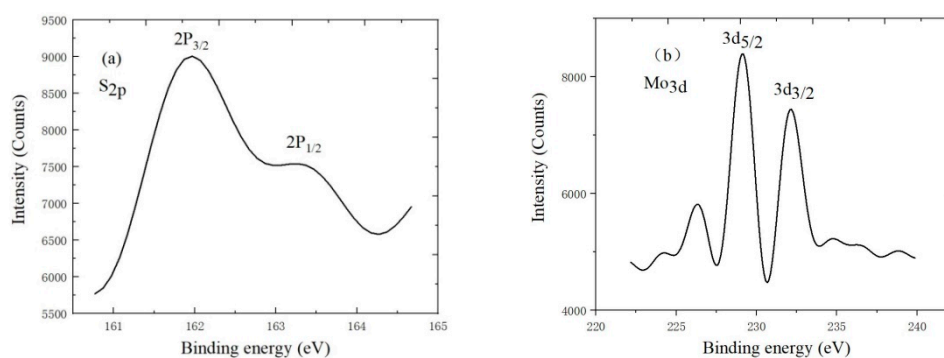


Figure 7. (a) S_{2p}; (b) Mo_{3d} of X-ray photoelectron spectroscopy.

4. Conclusions

In this paper, large-area high-quality uniform triangle monolayer MoS₂ is successfully grown on an SiO₂/Si substrate with oxygen plasma treatment and graphene quantum dot solution by APCVD. Effects of substrate processing conditions, such as oxygen plasma treatment time, power, and the amount of graphene quantum dot solution on growth quality and the area of monolayer MoS₂ are studied in detail. The optimal dosage of 1mg/ml graphene quantum dot solution is 20 μL, and the optimal power and time of SiO₂/Si substrate processing are 500 W and 90 s, respectively. Analysis and characterization of monolayer MoS₂ are carried out by Optical Microscopy, AFM, XPS Raman, and Photoluminescence Spectroscopy. The results show that the grown sample on the SiO₂/Si substrate under optimal cleaning conditions is a large-area high-quality, uniform, triangular monolayer MoS₂ with a side length of 200 μm, and it is very effective to treat SiO₂/Si substrate by oxygen plasma and graphene quantum dot solution, which can pave the way for the fabrication of optoelectronic devices.

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Conflicts of Interest: The authors declare no conflict of interest.

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