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Growth of highly uniform 2-inch MoS₂ wafers using liquid precursor spraying

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With the progress of study, MoS₂ has been proven to show excellent properties in electronics and optoelectronics, which promotes the fabrication of future novel integrated circuits and photodetectors. However, highly uniform wafer-scale growth is still in its early stage, especially regarding how to control the precursor and its distribution. Herein, we propose a new method, spraying the Mo precursor, which is proven to fabricate highly uniform 2-inch monolayer MoS₂ wafers. The Mo-precursor concentration and spray time are the key parameters, which have been systematically studied. The monolayer and bilayer coverage, Raman vibration, and PL emission properties are investigated. It was found that when the Mo-precursor concentration is 10 mg mL⁻¹ and spray time is 8 min, the as-grown MoS₂ wafer has the highest quality and electrical performance. By studying the electrical properties of transistor arrays, it was found that the MoS₂ transistors show slight vibration, the average ON/OFF ratio is 1.21×10^6 , and the maximum carrier mobility is 13.39 cm² V⁻¹ s⁻¹ without further optimizing the device fabrication. These results directly indicate that the spray method could fabricate MoS₂ wafers with both high optical and electrical uniformity. Moreover, the influence of bilayer coverage on the optical and electrical properties is studied, which demonstrates that additional bilayer nucleation would increase the scattering centers and thus suppress the electrical performance. By this method, we have successfully grown more than 100 2-inch wafers with a stable process, which further proves its potential application in future MoS₂ electronics and integrated circuits.

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Introduction

Two-dimensional materials have emerged as critical candidates for next-generation electronics due to their unique characteristics of atomic-scale thickness and modifiable band structures, which enable the fabrication of sub-nanometer narrow transistors and facilitate the construction of nanoelectronics for future post-Moore era.^{1–3} Up to now, although the 2D family has many members, molybdenum disulfide (MoS₂) has received the most investigations, including its growth and application in electronic devices and optoelectronic devices.^{4–6} A MoS₂-based integrated circuit has become more and more urgent. Recently, many industries have also paid attention to MoS₂, such as TMC, Samsung, *etc.* After more than ten years, many novel devices have been proposed and proven; however, controlled wafer-scale growth, the key factor in promoting MoS₂ in real applications, is still in its early stage. The uniformity in a wafer and wafer-to-wafer is hard to control.

Among the many growth methods, metal–organic chemical vapor deposition (MOCVD) and chemical vapor deposition (CVD) have become the dominant methods to grow MoS₂.^{7–14} In 2015, the Park group reported the growth of MoS₂ wafers by using Mo(CO)₆ and H₂S as the precursors.¹⁵ Due to the slower growth speed, it takes about one week for 2-inch growth. On the other hand, the normal CVD using evaporated Na₂MoO₄ has the best advantage of fast growth speed, which can even reach 144 μm min⁻¹.¹⁶ However, understanding how to control the uniform distribution of the precursor and further control the uniformity of the as-grown MoS₂ is important. More and more scientists are paying attention to this point. Zhang's group employed pre-oxidized Mo foil to replace MoO₃ or (NH₄)₂MoO₄ powder.¹⁷ It was found that MoS₂ can be grown as large as 6 inches. The key factor is controlling the uniformity of the pre-oxidized Mo foil. Apart from this method, although there are many literature studies that report large-scale growth, the methods are still not very good. A breakthrough was achieved by Wang *et al.* through a multi-source CVD configuration enabling a uniform point-source precursor reaction across 4-inch growth substrates.¹⁸ To enhance morphological homogeneity and operational stability in MoS₂ wafer fabrication, researchers developed a face-to-face deposition approach.

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This methodology employs precursor surface diffusion mechanisms through controlled vapor-phase delivery, achieving optimized spatial distribution *via* interfacial energy minimization. Notably, Tang's team engineered a dual-source deposition system by spin-coating S/Se-metal salt composites on molybdenum foil, achieving wafer-scale $\text{MoS}_{2(1-x)}\text{Se}_{2x}$ monolayers through controlled parallel substrate alignment.¹⁹ Concurrently, Chiu *et al.* demonstrated a stacked CVD approach with constant precursor gradients, realizing synthesis of monolayer MoS_2 with 490 cm^2 deposition coverage.²⁰ Despite these advancements, current face-to-face strategies exhibit intrinsic limitations in scalability and process economics, impeding industrial-scale manufacturing of 2D materials. This technological gap necessitates the creation of streamlined synthesis protocols that simultaneously optimize production efficiency, uniformity preservation, and thermodynamic stability. The realization of such synthesis protocols would accelerate the technological maturation of 2D semiconductors for next-generation optoelectronic devices and high-sensitivity sensor arrays.

In this work, we propose a novel face-to-face growth strategy for synthesizing wafer-scale monolayer MoS_2 on sapphire substrates. The method involves spraying a Mo precursor onto high-temperature-resistant quartz glass, which serves as a surface source. This approach ensures a uniform distribution of the Mo precursor across a carrier substrate with dimensions identical to those of the growth substrate, thereby significantly improving the uniformity and stability of the as-grown MoS_2 . Systematic investigations were conducted to evaluate the effects of Mo-precursor concentration and spray time on MoS_2 synthesis. Notably, when monolayer coverage reaches 100%, bilayer coverage exhibits a proportional increase with elevated precursor concentrations or extended spray times. Raman spectra characterization and PL spectra characterization of the as-grown MoS_2 reveal that bilayer nucleation critically influences the quality. Moreover, excessive bilayer nucleation density results in severe degradation of electrical properties, primarily due to bilayer nuclei acting as scattering centers. This scalable preparation method demonstrates simplicity and cost-effectiveness, enabling the fabrication of highly uniform and stable MoS_2 films for industrial applications.

Results and discussion

Since the growth of MoS_2 in 2010, more and more investigations have focused on how to grow large-scale MoS_2 films with high uniformity, thus enabling further potential applications in electronics and optoelectronics.^{21,22} Many efforts have been made, such as using MoO_3 and $(\text{NH}_4)_2\text{MoO}_4$ powder as the precursors, whose vaporization points are very close to the MoS_2 film growth temperature.^{23,24} However, the powder-like source leads to non-uniform evaporation, especially when it is used for large-scale growth. The non-uniform distribution of evaporation makes the MoS_2 film non-uniform, hindering its further application. Recently, pre-oxidized Mo foil with a

large area was placed face-to-face on a glass substrate.¹⁷ It was found that the as-grown MoS_2 can enlarge to 6 inches. Our group has also performed this experiment; however, it was found that the MoO_3 evaporation strongly depends on the state of the pre-oxidized Mo foil. To make the precursor stable and uniform, here, we propose a new method, namely spraying of Na_2MoO_4 solution, which can form a highly uniform Mo-precursor layer on 2-inch quartz glass, as shown in Fig. 1a. The uniformly distributed Na_2MoO_4 will react with the 2-inch quartz substrate to produce a Na–Mo–Si–O composite and then release a uniform molybdenum source.²⁵ After reaction with the S vapor released from the ZnS crystal, a uniform MoS_2 film is formed on the sapphire substrate, and the Zn element is not doped into MoS_2 (Fig. S1).²⁶ As shown in Fig. 1b, c and S2a–f, one can see that the MoS_2 film has similar contrast, indicating its uniformity. The step height in Fig. 1c is about 0.81 nm, which is very close to the intrinsic thickness of monolayer MoS_2 .²⁷ The high-resolution HAADF image shown in Fig. 1d exhibits a monolayer sandwich structure, further proving that the as-grown MoS_2 is a monolayer.²⁸ To further check the film microstructure, the line scans along the X and Y directions across the whole wafer are shown in Fig. 1e and f. As seen, the positions and intensities of the A_{1g} and E_{2g} bands are similar without typical variation, suggesting its high uniformity. PL spectra of the MoS_2 wafer could also reflect the uniformity in different regions, as shown in Fig. S3a and b. X-ray photoelectron spectroscopy was employed to study the as-grown MoS_2 film. As shown in Fig. 1g and h, one can see that the signal from 240 eV to 223 eV can be deconvoluted into Mo $3d_{5/2}$ and Mo $3d_{3/2}$, MoS_xO_y and S 2s. The existence of MoS_xO_y located at about 236.32 eV is caused by surface oxygen adsorption, which is widely observed in the literature.^{29,30} The S 2p spectra can be deconvoluted into S $2p_{1/2}$ and S $2p_{3/2}$. By normalizing the Mo and S areas in the XPS spectrum, it was found that the atomic ratio of S/Mo is about 1.89. This indicates the existence of sulfur vacancies.

In order to gain insight into the growth mechanism of Mo-precursor spraying, we first studied the influence of precursor concentration, as shown in Fig. 2. We fixed the spray time as 8 min and tuned the Mo-precursor concentration from 5 mg mL^{-1} to 15 mg mL^{-1} . The digital images of MoS_2 wafers (Fig. 2a–e) show the high coverage of the as-grown MoS_2 on sapphire. The monolayer coverage is 100% when the concentration of the aqueous solution of Na_2MoO_4 is increased in the range from 7.5 mg mL^{-1} to 15 mg mL^{-1} . As illustrated in Fig. 2f, when the Mo-precursor concentration is 5 mg mL^{-1} , the MoS_2 film is discontinuous, where the monolayer coverage is about 94.32% and no bilayer MoS_2 is formed. When increasing the Mo-precursor concentration, the coverage of bilayer MoS_2 increases. The bilayer MoS_2 coverage is less than 2% when the Mo-precursor concentration is in the range from 7.5 mg mL^{-1} to 10 mg mL^{-1} , which means that the MoS_2 film is a large-area uniform monolayer. The bilayer MoS_2 coverage can even reach about 10.32% when the precursor concentration is 15 mg mL^{-1} . Fig. 2g and h show that with the increase of Mo-precursor concentration, the Raman peak position of E_{2g} red shifts, while the A_{1g} position does not show

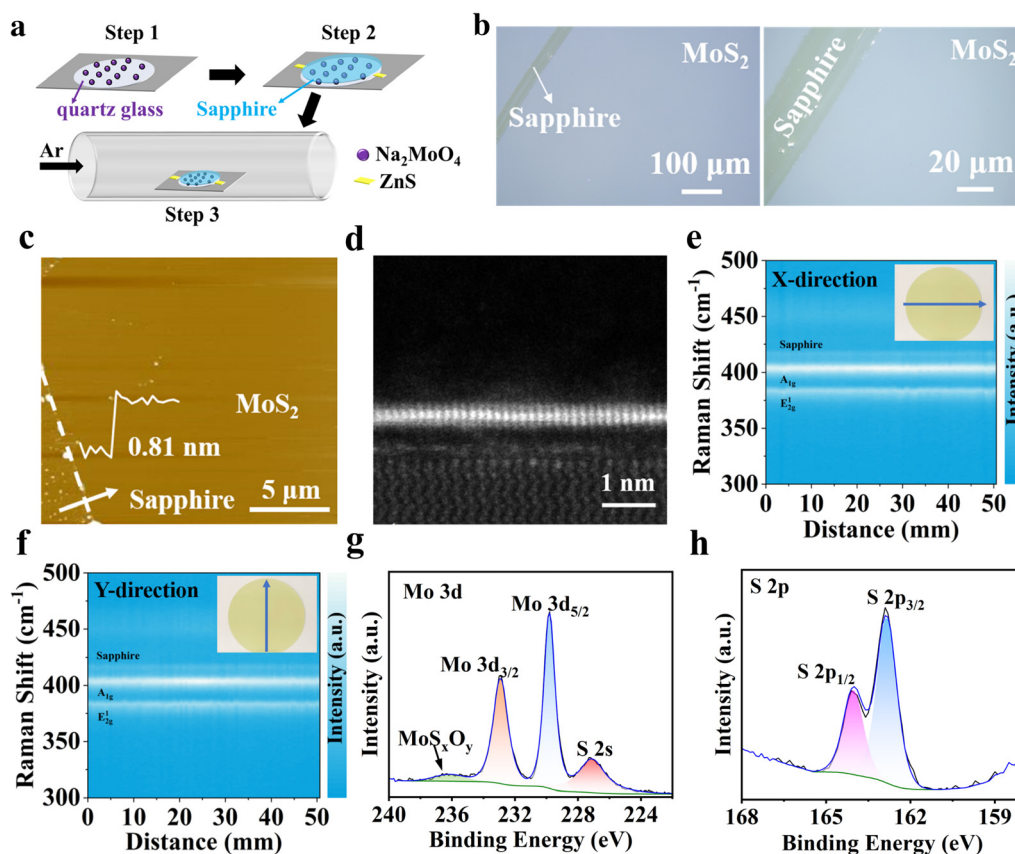


Fig. 1 The growth of a wafer-scale MoS₂ monolayer. (a) Diagram of the growth process. (b) The optical image of the MoS₂ film on sapphire. (c) The AFM image. (d) The high-resolution HAADF image of the section. (e and f) The Raman spectrum along the X and Y directions across the 2-inch wafer. (g and h) The high-resolution Mo 3d and S 2p spectra.

obvious change. The peak position difference ($A_{1g}-E_{2g}$) is well known to correlate with the thickness of MoS₂.³¹ As seen, with increasing Mo-precursor concentration, the value of $A_{1g}-E_{2g}$ changes from 17 cm⁻¹ to 21 cm⁻¹, which is well consistent with the evolution of bilayer MoS₂ coverage. The PL spectra are shown in Fig. 2i. It can be seen that when the Mo-precursor concentration is 7.5 mg mL⁻¹ or 10 mg mL⁻¹, the PL peak centers are located at 1.88 eV, which is well consistent with exfoliated MoS₂ single-crystal flakes.³² However, when the Mo-precursor concentration is less than 7.5 mg mL⁻¹ or larger than 10 mg mL⁻¹, the PL peak centers all redshift suggesting that the optical bandgap becomes lower, which may be caused by the interaction between the bilayer and the monolayer. Statistical analysis of the PL spectra (Fig. S4a) clearly exhibits a sharp decrease of PL intensity and an evident increase of full width at half maxima (FWHM) when increasing the Mo-precursor concentration in the range from 7.5 mg mL⁻¹ to 15 mg mL⁻¹. Combining the Raman vibration analysis and PL emission analysis, one can conclude that the MoS₂ film grown using a 7.5 mg mL⁻¹ concentration has the highest quality.

Besides the precursor concentration, the spray time is also an important factor, which has been systematically studied as shown in Fig. 3. The Mo-precursor solution is fixed at 7.5 mg

mL⁻¹, and the spray time is increased in the range from 7 min to 11 min. As shown in Fig. 3a–e, the growth of monolayer MoS₂ is continuous and has extremely high coverage of 100% when the spray time is more than 7 min. Moreover, the spray time and bilayer coverage are positively correlated, where a longer spray time will lead to higher bilayer coverage. As illustrated in Fig. 3f, the monolayer coverage is 100% and the bilayer coverage is lower than 1% while spraying for 8 min. Raman spectra of MoS₂ with different spray times are shown in Fig. 3g. The statistical analysis of Raman spectra (Fig. 3h) indicates that the peak of E_{2g} exhibits a redshift and the value of $A_{1g}-E_{2g}$ changes from 17 cm⁻¹ to 21 cm⁻¹ with extending the spray time. The increased bilayer coverage can enhance the interlayer interaction forces between the monolayer and bilayer.³³ PL spectra of MoS₂ with different spray times (Fig. 3i) indicate that their optical bandgap is about 1.8–1.9 eV, well consistent with the nature of monolayer MoS₂.³⁴ As illustrated in Fig. S4b, the statistical analysis of the PL spectra demonstrates that the PL intensity rises and FWHM broadens when the spray time is longer than 8 min. Combining the Raman spectra analysis and PL spectra analysis, it can be concluded that the quality of the MoS₂ film is relatively high when the spray time is 8 min and the Mo-precursor concentration is

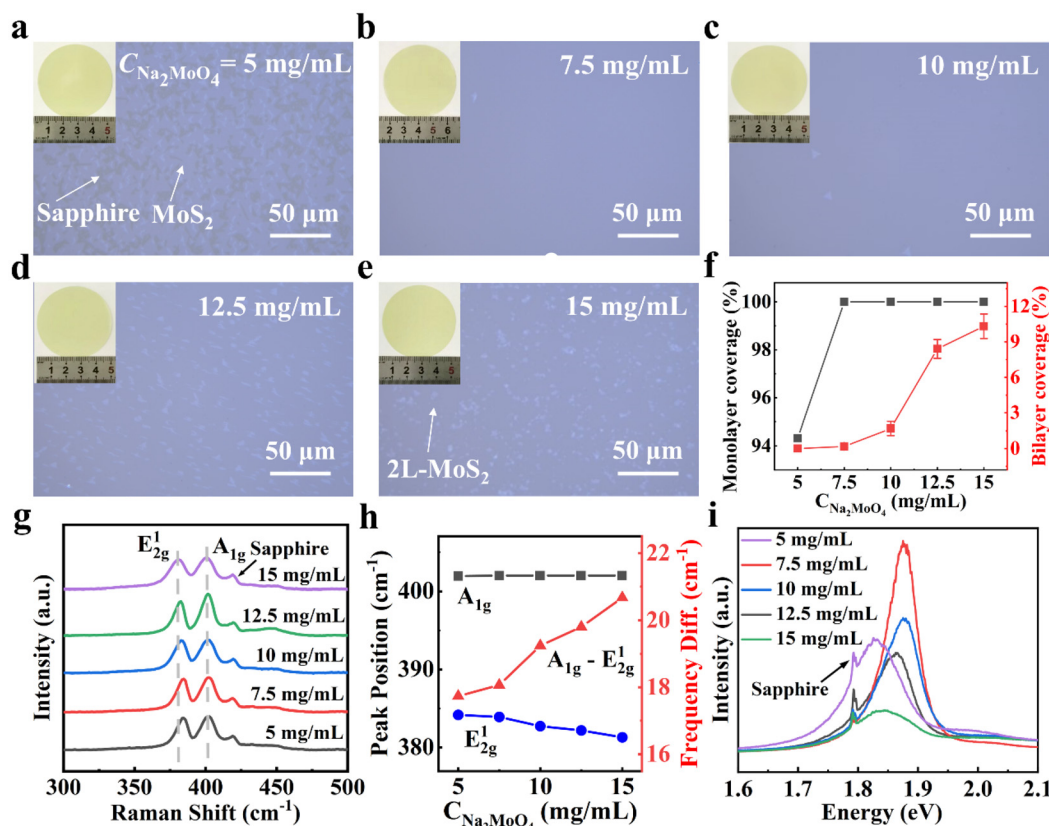


Fig. 2 The influence of Mo-precursor concentration on MoS₂ growth. (a–e) The digital MoS₂ wafer images and the optical microscopic images with different Mo-precursor concentrations. (f) The monolayer coverage and bilayer coverage as a function of Mo-precursor concentration. (g) Raman spectra of MoS₂ with different Mo-precursor concentrations. (h) The peak positions of E_{2g} and A_{1g} and the frequency difference of A_{1g}–E_{2g} as a function of Mo-precursor concentration. (i) PL spectra of MoS₂ with different Mo-precursor concentrations.

7.5 mg mL⁻¹. In addition, as shown in Fig. S5 and S6, the growth temperature and environmental pressure have also been studied. It was further determined that 100% monolayer of MoS₂ was obtained at a growth temperature of 820 °C and a pressure of 1 Torr, and there was almost no bilayer nucleation coverage. By this method, we have successfully grown 100 2-inch wafers with a stable process, as illustrated in Fig. S7.

According to the above study, it can be found that with increasing the Mo-precursor concentration and spray time, the bilayer coverage increases while the monolayer coverage reaches 100% on sapphire, which provides a platform to study the optical property evolution from the monolayer to the bilayer. The influence of bilayer coverage on the characterization of Raman and PL spectra is summarized in Fig. 4. As shown in Fig. 4a, the Raman peak position of E_{2g} presents a trend of redshift, and the value of A_{1g}–E_{2g} increases with the bilayer coverage increase. This may be attributed to defects such as sulfur vacancies and lattice dislocations that cause changes in the in-plane lattice stretching state, resulting in a redshift of E_{2g} while A_{1g} remains unchanged.³⁵ Furthermore, the increase of bilayer coverage may lead to increased inter-layer interactions, which also contributes to the increase of A_{1g}–E_{2g}.³³ The PL energy and FWHM as the function of bilayer

coverage are illustrated in Fig. 4b. The FWHM has a negative correlation with bilayer coverage. The PL peak of the direct bandgap red shifts with increasing bilayer coverage, exhibiting that bilayer coverage can change the surface state of the MoS₂ film, affecting the surface electronic state and band structure and leading to the decrease of the direct bandgap peak binding energy.³⁶ Further pretreatment, such as pre-annealing the sapphire substrate, may improve the surface quality and then suppress bilayer nucleation and bilayer coverage.³⁷

To investigate the influence of Mo-precursor spraying on electrical properties of MoS₂ films, transistor arrays were constructed based on MoS₂ films with different Mo-precursor concentrations and spray times, as shown in Fig. 5a. The film is transferred from the growth substrate to a fresh SiO₂/Si substrate under the assistance of PMMA. In order to remove oxygen and other adsorbed impurities on the surface of the MoS₂ film, the transistor arrays were annealed at 250 °C for two hours under an Ar/H₂ atmosphere. The transfer curves and typical output curves (Fig. 5b, c and Fig. S8–S11) indicate that the MoS₂ film grown by using 10 mg mL⁻¹ Na₂MoO₄ solution and spraying for 8 min has the best homogeneity and electric-field effect. The histograms of the carrier mobility and the I_{on}/I_{off} ratio, as illustrated in Fig. 5d, e and Fig. S12–S14, show that

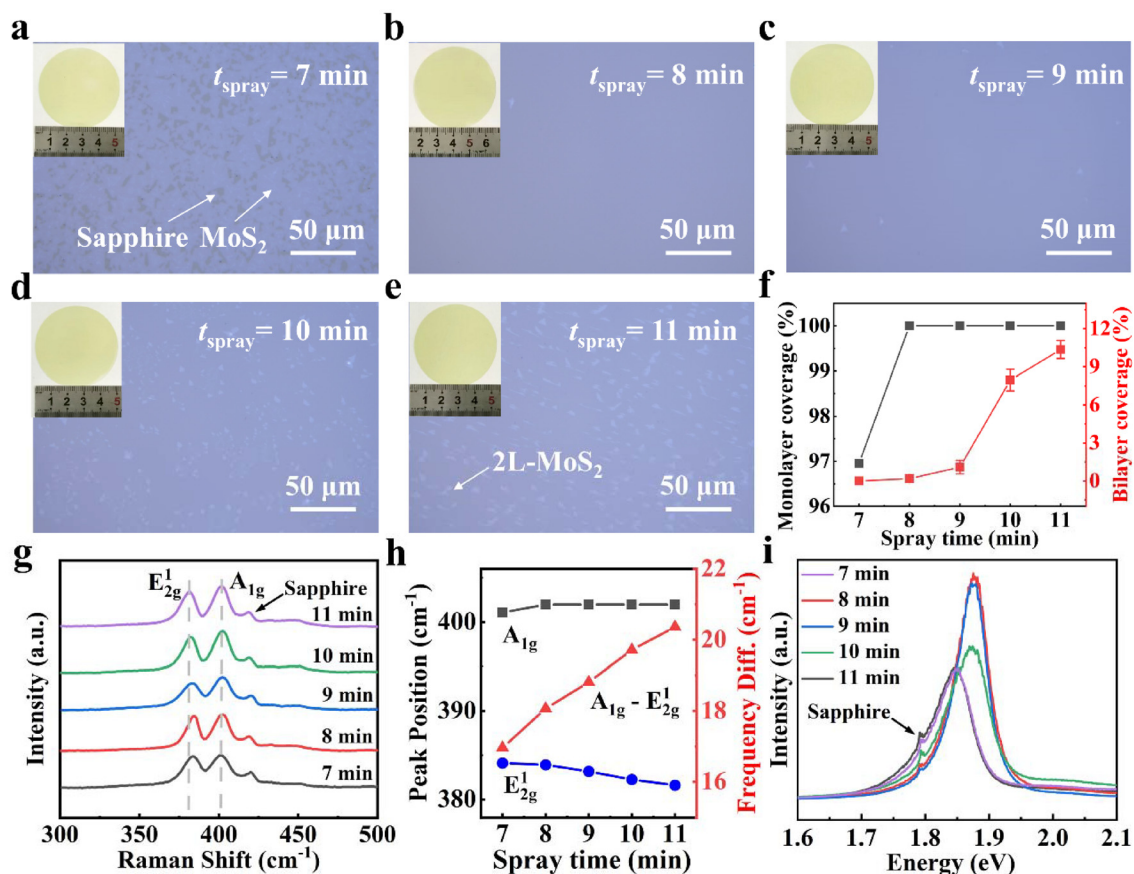


Fig. 3 The influence of spray time on MoS₂ growth. (a–e) The digital MoS₂ wafer images and the optical microscopic images with different spray times. (f) The monolayer coverage and bilayer coverage as a function of spray time. (g) Raman spectra of MoS₂ with different spray times. (h) The peak position of E_{2g}, A_{1g} and the frequency difference of A_{1g}–E_{2g} as a function of spray time. (i) PL spectra of MoS₂ with different spray times.

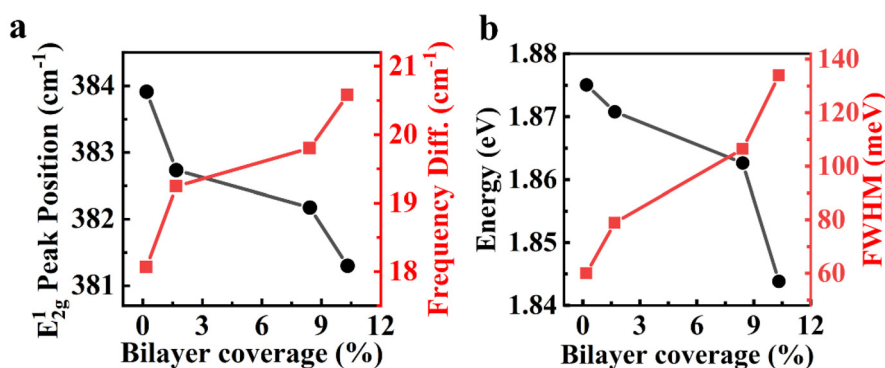


Fig. 4 The influence of bilayer nucleation coverage on the optical properties. (a) The peak position of E_{2g} and frequency difference of A_{1g}–E_{2g} as a function of bilayer coverage. (b) The PL energy as a function of bilayer coverage.

the Mo-precursor concentration and the spray time critically govern the electrical properties of MoS₂ films. Using 10 mg mL⁻¹ Na₂MoO₄ aqueous solution and a spray time of 8 min, the MoS₂ carrier mobility can reach 13.39 cm² V⁻¹ s⁻¹ ($\mu = g_m \frac{L}{WC_g V_{ds}}$ is used to extract mobility from transfer curves) and the I_{on}/I_{off} ratio is 1.21×10^6 , and the average

values of carrier density, interface trap density and subthreshold swing are 1.77×10^{12} cm⁻², 3.46×10^{12} eV⁻¹ cm⁻² and 2.89 V dec⁻¹, respectively (Fig. S15). The interface trap density and subthreshold swing of the grown MoS₂ are not much different from the values reported in the literature.³⁸ It should be noted that carrier mobility is slightly lower compared with the highest value reported in the literature; this should be

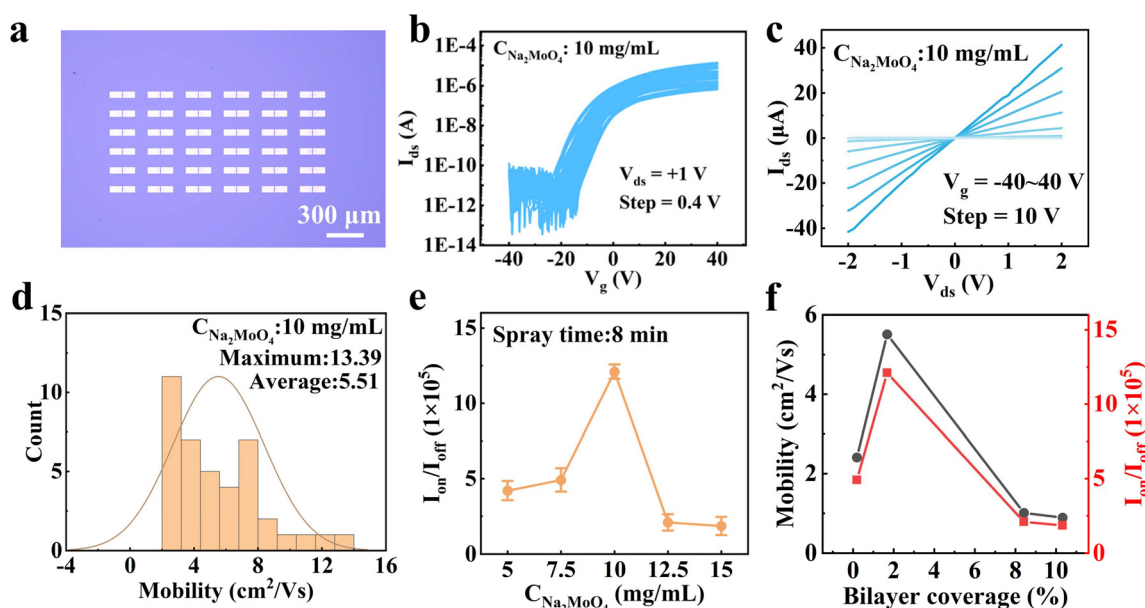


Fig. 5 The electrical properties of the MoS₂ film. (a) The optical image of the transistor array, where the channel length is 5 μm and the channel width is 20 μm. (b) The transfer curves. (c) The typical output curves. (d) The histogram of the carrier mobility where the MoS₂ film is grown by using 10 mg mL⁻¹ Mo-precursor concentration and spraying for 8 min. (e) The $I_{\text{on}}/I_{\text{off}}$ ratio as a function of Mo-precursor concentration. (f) The carrier mobility and $I_{\text{on}}/I_{\text{off}}$ ratio as a function of bilayer coverage.

caused by the contact electrode properties and dielectric surface trapping effects.^{39–41} As shown in Fig. S16, the carrier mobility of MoS₂ grown by CVD is consistent with that of mechanically exfoliated MoS₂, which further indicates the high quality of the film. And the high value of SS in our device is probably attributed to the dielectric interface, such as the surface state and charge traps.⁴² Further optimization, including the dielectric interface and contact engineering, will facilitate the improvement of the device performance.^{43–45} Furthermore, the carrier mobility and $I_{\text{on}}/I_{\text{off}}$ ratio as a function of bilayer coverage are plotted in Fig. 5f. It shows that both of these factors show largest values when the bilayer coverage is 1.68%. In our case, lower bilayer coverage may suggest that there are some point defects formed since the Mo precursor is not enough. However, more Mo precursors will lead to bilayer nucleation and cause Fermi fluctuation and grain boundaries, resulting in suppression of the electrical performance.

Conclusions

In summary, we proposed a new method, Mo-precursor spraying, which can facilitate the growth of highly uniform monolayer MoS₂ wafers. The influence of spraying on MoS₂ preparation was studied by optimizing the Mo-precursor concentration and spray time. This work demonstrates that the Mo-precursor concentration and spray time critically govern the continuity and uniformity of monolayer MoS₂ films, which exhibit a positive correlation with bilayer coverage. Additionally, the effects of bilayer coverage on Raman spectra,

PL spectra and electrical properties of MoS₂ films are studied. As bilayer coverage increases, the Raman peak position of E_{2g} red shifts and A_{1g}–E_{2g} increases. The PL peak position, correlating with the bandgap, shows a trend of redshift and its FWHM becomes broad. Furthermore, high-density and small-sized bilayer nuclei would act as scattering centers, leading to serious deterioration of the electrical properties of the monolayer film. Combining the Raman spectra analysis, PL spectra analysis and electrical measurements, the MoS₂ film grown by using a 10 mg mL⁻¹ Na₂MoO₄ solution and spraying for 8 min exhibited relatively high crystal quality. An average $I_{\text{on}}/I_{\text{off}}$ ratio of 1.21×10^6 and a maximum carrier mobility of 13.39 cm² V⁻¹ s⁻¹ were observed. This simple and cost-effective preparation method provides a general technical means for the wafer-scale growth of 2D materials.

Experimental section

CVD growth of MoS₂ monolayer films

The surface of high-temperature resistant quartz glass was spray-coated with 7.5 mg mL⁻¹ Na₂MoO₄ aqueous solution for 8 min. The parameters of spraying are as follows: the area of the circular nozzle is 7.07 cm², the distance between the nozzle and the substrate is 8 cm, the velocity of the sprayed jet is about 0.2 mL min⁻¹, and the source pressure is 80 kPa. A sandwich structure made of sapphire/ZnS/quartz glass is placed at the center of the tube furnace, and ZnS with a size of 5 mm × 5 mm is inserted into the edge between the sapphire and the quartz glass, which not only provides the active sulfur monomer but also adjusts the distance between the sapphire

and the quartz glass by varying its thickness. 400 sccm Ar was introduced into the tube furnace as the carrier gas. The growth process was then conducted at a growth temperature of 800 °C under a vacuum pumped to 1 Torr for 30 minutes. After the growth was completed, the tube furnace was allowed to cool naturally.

Characterization methods

The optical images were obtained using SG-9XF. AFM was measured using MultiMode-8, Bruker. Raman and PL spectra were recorded using HORIBA with an excitation wavelength of 532 nm. HAADF was performed using Talos F200X with the aberration-corrected function. XPS was performed on AXIS SUPRA+.

Device fabrication and measurements

First, the fabrication of MoS₂ transistor arrays commenced with the preparation of source/drain electrodes on a Si/SiO₂ substrate, where a uniform film of the NR9-3000PY negative photoresist was first spin-coated (4000 rpm, 20 s) and pre-baked at 140 °C for 7 min to remove solvent and enhance adhesion. Subsequent patterning was achieved using a contact aligner with a 10 s exposure time, followed by post-exposure baking (140 °C, 7 min) to harden the resist, development in an AZ 400K developer for 45 s to form the template, electron-beam evaporation of a Ti/Au bilayer (10 nm Ti/15 nm Au), and final removal of residual photoresist using acetone and isopropanol. Second, an 8% polymethyl methacrylate solution (PMMA) was spin-coated (4000 rpm, 50 s) onto the MoS₂-grown sapphire substrate to form a support layer, which was annealed on a hotplate at 180 °C for 3 min to strengthen interfacial adhesion. Leveraging the hydrophobicity contrast between sapphire and PMMA, the MoS₂/PMMA stack was then completely delaminated *via* a deionized water flotation method and transferred onto the prefabricated source/drain electrodes. After vacuum-drying for 1 h, the PMMA support was dissolved by sequential rinsing in acetone and isopropanol, resulting in an MoS₂ channel layer aligned with the metal electrodes. In the end, to define the channel region, micropatterning was performed using the RZJ-304 positive photoresist: spin-coated (4000 rpm, 20 s), soft-baked on a hotplate (100 °C, 5 min) to ensure uniformity, exposed *via* contact lithography (8 s), developed in an AZ 400K developer for 25 s to form an etch mask, selectively etched using an oxygen plasma system to remove excess MoS₂ while preserving the channel structure with a width of 20 µm and a length of 5 µm, and finally, the photoresist was stripped with acetone, thus completing the MoS₂ transistor array fabrication. The electrical measurements were performed using an LPS-100 vacuum probe station with Keithley 2614 source meters.

Author contributions

Z. W. conceived, designed and supervised the project. X. L., J. L., D. L., C. Z., X. Z., Z. W. and C. T. conducted the experi-

mental work and performed the data analysis. X. L. and Z. W. wrote the manuscript. All authors contributed to the discussion of the results and editing of the manuscript.

Conflicts of interest

There are no conflicts to declare.

Data availability

The data supporting this article have been included as part of the SI.

Acknowledgements

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