



High-Performance Field-Effect Transistor Fabricated on CVD-Grown MoS₂ Monolayers with Indium Contacts

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Abstract

Molybdenum disulfide (MoS₂), an emerging two-dimensional semiconductor material, has been keenly studied for field-effect transistors (FETs). In this work, we explored the optical and electrical properties of FETs fabricated by MoS₂ flakes grown by chemical vapor deposition (CVD) and transferred to the electrodes through propylene carbonate film. Large-area, high-quality and highly crystalline MoS₂ monolayers up to 58 μm are obtained through CVD. Flakes are characterized by optical microscopy, atomic force microscopy, Raman spectroscopy, and photoluminescence analysis. The back-gated measurements are performed in ambient conditions without any encapsulation of the device. The fabricated device reveals *n*-type behavior with high field-effect mobility of 32 cm²/V s and high current ON/OFF ratio of 10⁶. Good ohmic contact is achieved while using indium as source/drain electrodes. The large sized, highly crystalline flakes of MoS₂ and the fabricated device showing high field-effect mobility and ON/OFF ratio make them potential candidates for high-performance nanoelectronics and optoelectronics devices.

Keywords Two-dimensional materials · MoS₂ · chemical vapor deposition · propylene carbonate · field-effect transistor · indium

Introduction

Because of its unique and remarkable electrical properties, graphene has potential for substitution in typical Si-based semiconductor devices.¹ Although graphene has extraordinary high carrier mobility,² its zero band gap limits its use in logic applications. Inspired by the invention of graphene monolayers, transition metal dichalcogenides (TMDs) such as MoS₂, MoSe₂, WS₂ and WSe₂ have gained significant consideration for their excellent potentials in research fields of nanotechnology, microelectronics, photonics, and optoelectronics.^{3,4} TMDs possess a layered hexagonal structure having a dangling bond-free surface, excellent thermal conductivity, and stability.⁵ Among these TMDs, MoS₂ has been

studied widely as it exhibits excellent flexibility, electrical and optical properties.⁶ In MoS₂, each Mo atom is covalently bonded to two sulfur atoms. This covalent bonding provides great mechanical strength to MoS₂. MoS₂ has an outstanding ON/OFF ratio of about 10⁸ and carrier mobility almost equal to 200 cm²/V s at standard temperature.⁷ MoS₂ has a tunable bandgap, from 1 to 2 eV.^{8,9} The bandgap of MoS₂ changed from an indirect to a direct one from bulk MoS₂ to monolayer MoS.⁹ Monolayer MoS₂ has a direct bandgap of 1.9 eV which makes it suitable for use in electronic and optoelectronic devices.¹⁰

Thin layers of MoS₂ can be obtained by various methods such as mechanical exfoliation,⁷ liquid exfoliation,¹¹ chemical vapor deposition (CVD),¹² sulfurization,¹³ physical vapor deposition,¹⁴ and hydrothermal¹⁵ and electrochemical synthesis.¹⁶ Flakes of high quality can be achieved by exfoliation, but these flakes are randomly dispersed, limiting control of the number of layers of MoS₂. Hence, these methods are not designed for large-scale device fabrication. By synthesis, MoS₂ forms nanotubes (one-dimensional) or closed (zero-dimensional) structures. It is very rare to obtain large-area, high-quality, layer-controlled and high-yield MoS₂ for bulk fabrication of devices with excellent

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electrical properties. CVD is used extensively to produce large-area layers of MoS₂ with control over the number of layers, but this method does not ensure the high quality of the flakes. Enhanced scalability has been reported by using various methods, for example sulfurization of molybdenum and its oxides, vapor-liquid growth by using powder MoO₃ and sulfur, thermal decomposition of ammonium thiomolybdates, and vapor-solid growth of MoS₂ monolayers by using powder of MoS₂.¹⁷ In emerging CVD methods, sulfurization is done after spin coating of a solution of metal on a substrate.¹⁸ Sulfurization with sulfur vapor has been demonstrated to be useful for fabricating sputtered-multilayer MoS₂ films with an approximately 4-nm thickness with enhanced mobility.¹⁹ Monolayers of MoS₂ have been achieved by CVD on a mica substrate. A 7.5-nm-thick MoS₂ film was produced. Except for monolayers, no other layers of MoS₂ have been fabricated. With the help of a nucleation promoter, perylene-3,4,9,10-tetracarboxylic acid tetrapotassium salt (PTAS), a highly crystalline and uniform monolayer of MoS₂ has been fabricated.²⁰ However, it is very hard to use this monolayer uniformly on a large substrate. Wafer-scale monolayers of MoS₂ have been synthesized which exhibit on/off ratios ranging from 10⁵ to 10⁸ and carrier mobility up to 1.2 cm²/V s.²¹ A recent work investigating field-effect transistors (FETs) fabricated on monolayer MoS₂ produced by CVD showed carrier mobility of 1.45 cm²/V s with an ON/OFF ratio of 10⁴.²² MoS₂ FETs fabricated by mechanical exfoliation have shown good carrier mobility, but FETs based on CVD-grown MoS₂ exhibited low electrical performance. Growing a layer-controlled, contamination-free, high-quality, large-scale MoS₂ films for nano-devices with excellent performance is still challenging. Moreover, no optimized parameters and methods have been reported for the growth of monolayer MoS₂.

In this work, we fabricated uniform, highly crystalline, large-area, high-quality monolayers of MoS₂ by CVD on

SiO₂ substrates. Atomic force microscopy (AFM), Raman spectroscopy, and photoluminescence (PL) analysis confirmed that our monolayers of MoS₂ are highly scalable, uniform, and crystalline in nature. We also fabricated FETs on CVD-synthesized monolayer MoS₂ films which exhibited an on/off ratio of 10⁶ with comparative carrier mobility of 32 cm²/V s. The contact between the metal electrodes and the MoS₂ becomes one of the most critical aspects restricting device performance. Good ohmic contact is attained when we use indium (In) as electrodes, which improves the performance of the MoS₂-based FETs. Our research work demonstrates that excellent electrical performance of devices based on MoS₂ can be achieved by controlled growth of large-area and high-quality MoS₂ films.

Experimental Setup

MoS₂ Synthesis

Figure 1 demonstrates the setup of the experiment. Thin films of MoS₂ are grown on O₂ plasma treated 100 nm SiO₂/Si substrates in 2-zone CVD with 5 cm quartz. The substrate was pre-treated with O₂ plasma for 120 seconds at 110 W to attain a hydrophilic surface. MoO₃ and sulfur powders purchased from Sigma-Aldrich are used as precursors. Two milligrams of MoO₃ powder was taken in a quartz boat with the substrate in a face-down position arranged on the top of boat. Next, 300 mg of sulfur powder was placed in a separate boat. A distance of 15 cm was maintained between boats containing the MoO₃ and sulfur powder. Sulfur was placed in an upstream position in the furnace. Thin layers of MoS₂ were grown at 750°C under 500 sccm Ar flow.

The shape, density, and growth of the MoS₂ flakes were observed by optical microscopy (OM). The uniformity,

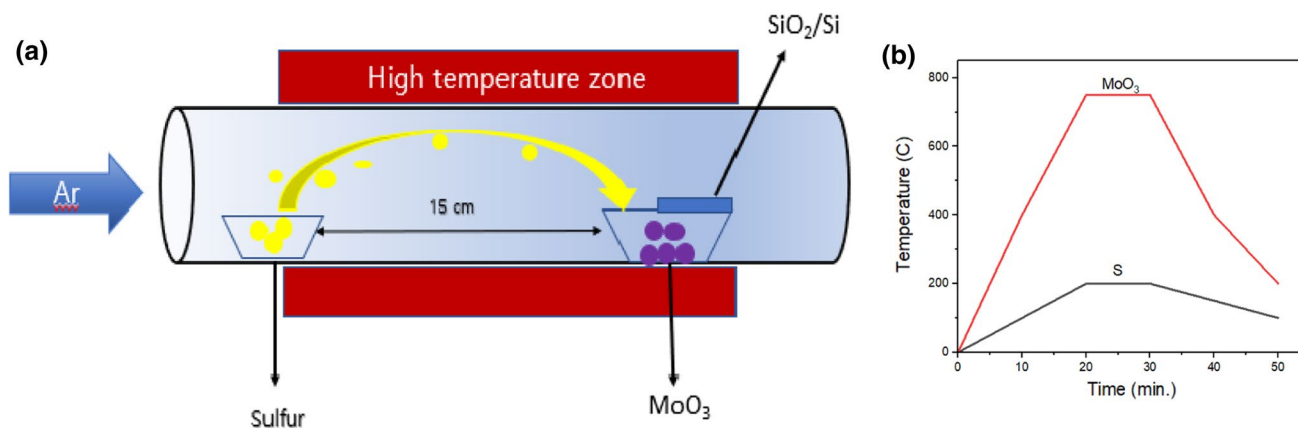


Fig. 1 (a) Schematic diagram of the CVD setup for growth of MoS₂. (b) Temperature profile of S and MoO₃ during growth process.

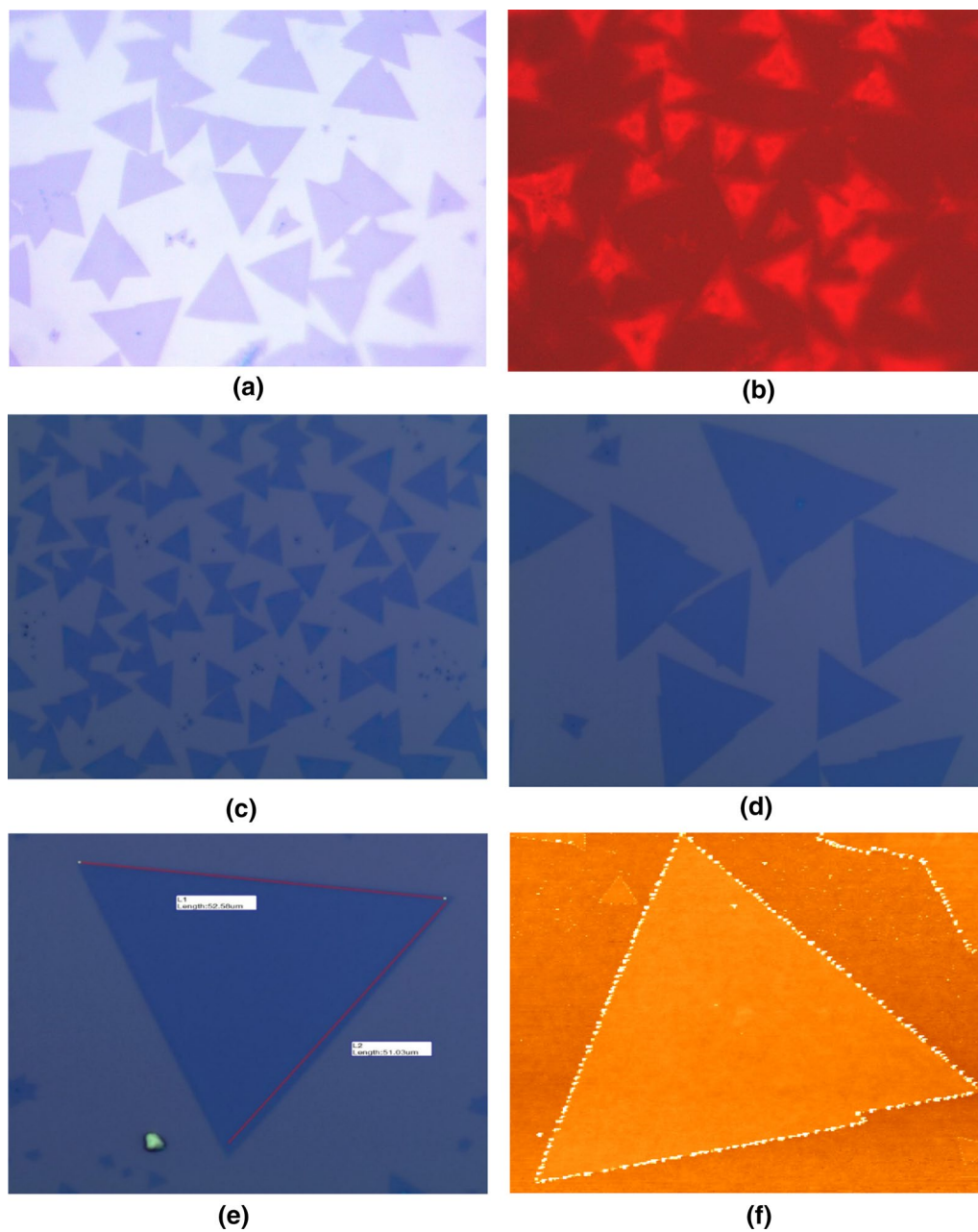


Fig. 2 (a) OM image of MoS₂ flakes. (b) PL image of MoS₂ flakes. (c, d) Density of MoS₂ monolayer flakes. (e) Area of MoS₂ monolayer flake. (f) AFM image of MoS₂ flake.

smoothness and surface structure were examined by a Nano-Magnetics ezAFM system. Raman and PL analysis of MoS₂ flakes were performed using a Renishaw inVia confocal Raman microscope system with an objective lens of 100× with numerical aperture of 0.75 A. continuous wave laser of 532 nm with power of 1 mW was used. Integration time of 0.3 s was used for Raman spectra and 0.02 for PL analysis.

Device Fabrication and Characterizations

The back-gated FET was fabricated by transferring the CVD-grown MoS₂ monolayer on the SiO₂/Si substrate, and Au/In metal electrodes were deposited on it by optical lithography. A total of 80 nm of Au was deposited on 20 nm indium (In) to make electrodes. The MoS₂ flakes synthesized via CVD were relocated onto a SiO₂/Si substrate using a pick-up technique involving the application of propylene carbonate (PC). The PC solution was made by combining

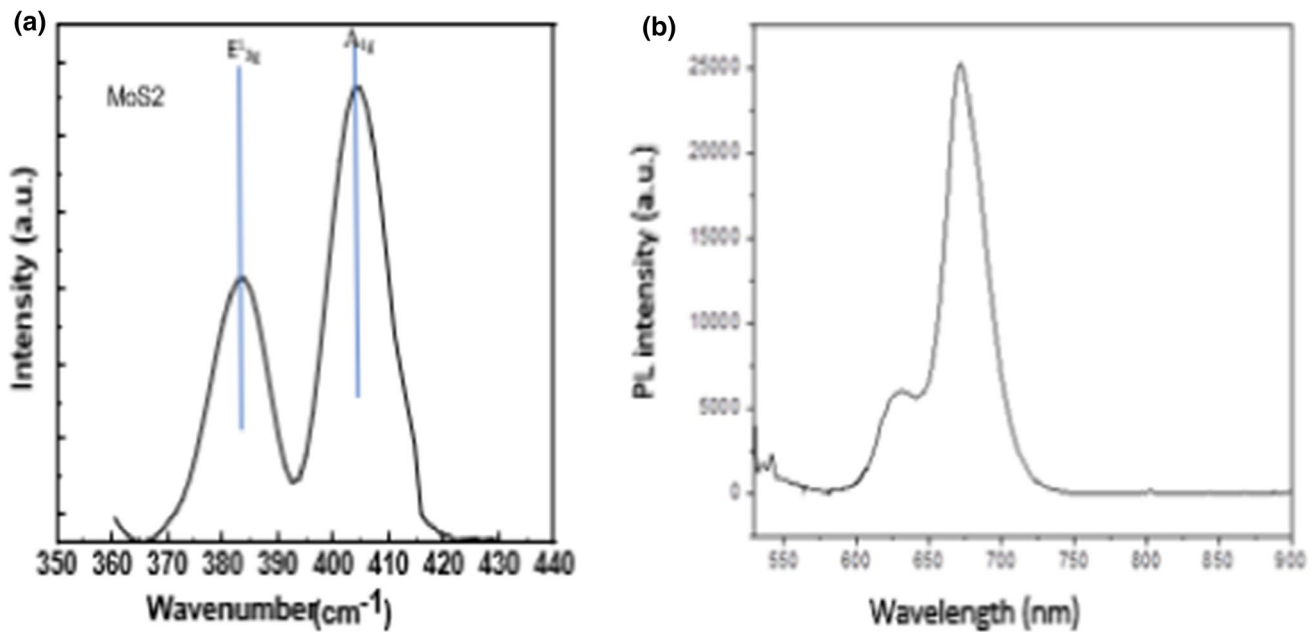


Fig. 3 (a) Raman spectra of MoS₂. (b) PL spectra of MoS₂.

methoxybenzene with PC particles at a mass fraction of around 10%. PC solution was drop-cast on the SiO₂/Si substrate containing CVD-grown MoS₂ flakes. The PC solution was hardened into the PC film by placing it on a hot plate for 10 min at 110°C. The PC film worked as a stamp and carried the MoS₂ layers out of the substrate. Then this PC film was moved to the targeted substrate. At 80°C, the PC film loosened its adhesion and MoS₂ layers were transferred on the desired substrate by removing the PC film. The transfer characteristics of the MoS₂-based FET were measured with a Keithley 2400 source meter.

Results and Discussion

Optical Characterization

Figure 2a presents the OM images of CVD-grown MoS₂ flakes with triangular shape. PL images of MoS₂ flakes can be seen in Fig. 2b. In Fig. 2c and d the density of the CVD-grown MoS₂ flakes can be seen. We observed maximum crystal size up to 58 μm for this growth. The OM image of a MoS₂ triangle with edges longer than 52.58 μm can be seen in Fig. 2e. In Fig. 2f, a smooth, uniform, highly crystalline and large-area layer structure of MoS₂ flake can be observed with AFM.

Figure 3a presents the Raman spectra of MoS₂. Most significantly, the Raman spectra of MoS₂ show two prominent peaks of MoS₂. Two non-resonant characteristic peaks of MoS₂ are found. These two peaks are dominated by two vibrational modes. E_{2g}^1 (in-plane vibration) is due to the in-plane vibration of two sulfur atoms with Mo atoms. The E_{2g}^1 mode appeared at 382 c/m. The A_{1g}^1 mode (out of plane) refers to the out-of-plane vibrations of sulfur atoms in opposite directions. The A_{1g}^1 mode is observed at 404 c/m. These two modes are very sensitive to the thickness of the MoS₂ structure. The thickness and number of layers of MoS₂ are determined by the peak positions of these two modes. The difference between these two peaks tells us about the number of layers of MoS₂. The difference between these two peaks, observed from our Raman spectra, is $A_{1g}^1 - E_{2g}^1 = 405 - 385 = 20$ c/m and ratio of $A_{1g}^1/E_{2g}^1 = 1.057 \approx 1$. The difference of 20 c/m shows that our MoS₂ is monolayer.^{23,24} Figure 3b presents the PL spectra of MoS₂. Excitons are the electron-hole pairs which are formed by optical excitation of electrons from valence band to conduction band when MoS₂ is exposed to laser of energy below than 670 nm. As these excited electrons are unstable, they come back from the conduction band to the valence band and relax. In the relaxation state, these electrons recombine with the holes produced in the valence band after their excitation. Radiation is induced because of this recombination. As a result, we observed various peaks of MoS₂ in its PL spectra. Two

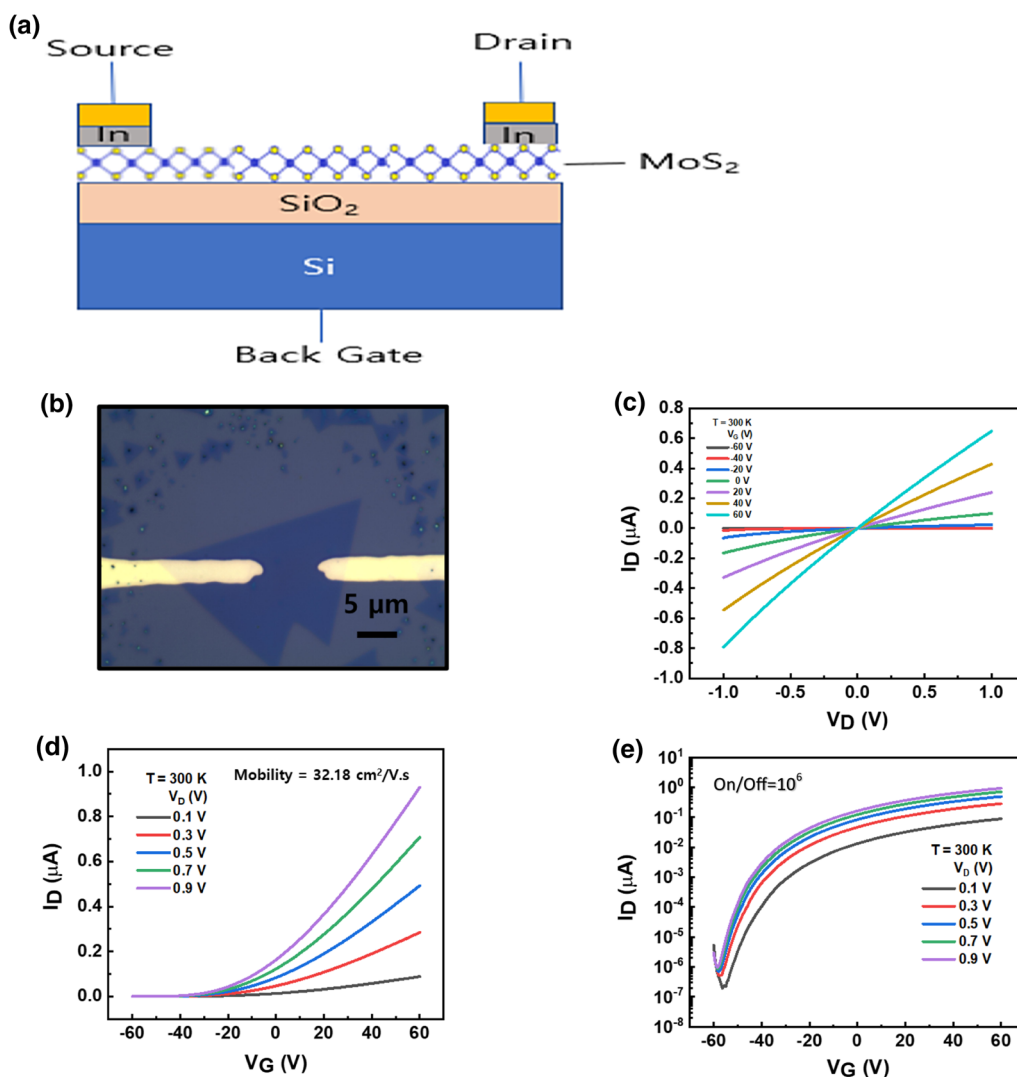


Fig. 4 (a) Cross-sectional diagram of CVD-grown MoS₂-based FET device. (b) Optical image of fabricated MoS₂-based FET. (c) Output curves of device by sweeping the V_G from -60 to 60 V. (d, e) Transfer characteristics (I_D - V_G) of monolayer MoS₂-based FET.

well-defined peaks are observed in bare MoS₂ spectra at 1.84 eV and 2.08 eV. These peaks are referred to as A exciton and B exciton, respectively. These peaks demonstrate the exciton formation, and this difference in energy between these two peaks is attributable to the spin-orbit splitting of the valence band.²⁵

Electrical Characterization

Figure 4a shows a cross-sectional illustration of the CVD-grown MoS₂-based FET device. The device is fabricated on a SiO₂/Si substrate. SiO₂ with a thickness of 300 nm acts as

the gate oxide layer deposited on 500-nm-thick Si. It acts as a back gate to control the fabricated FET. The channel length L_C of the active region is 5 μm and channel width W_C is 6 μm . Figure 4b shows the optical image of the fabricated MoS₂-based FET. Figure 4c, d demonstrates the output and transfer characteristics of the monolayer MoS₂-based FET in contact with In. Output curves of the FET were obtained by taking the V_g from -60 V to 60 V. Good ohmic contact with In was achieved. All electrical measurements were taken in ambient conditions. It is obvious from the transfer curve in Fig. 3d that MoS₂-based FET exhibits n -type behavior. From the saturation region of the on-state conducting channel, threshold voltage V_{TH} is defined. V_{TH} is obtained from

the intercept made from linear extrapolation on the I_D - V_G graph. FET is usually in the on state when $V_G=0$ V, which represents the collective conduction of electrons. Figure 3e represents the I_D - V_G curves demonstrating the distinct n -type characteristics with ON/OFF = 10^6 .

The field-effect mobility of the MoS₂ -based FET is obtained by using the following equation.

$$\mu_{\text{eff}} = \left(\frac{L}{C_{\text{ox}} W V_{\text{ds}}} \right) \left(\frac{dI_{\text{ds}}}{dV_{\text{gs}}} \right)$$

where μ_{FE} is field-effective mobility, g_m is transconductance = $\frac{\partial I_D}{\partial V_{\text{gs}}}$, L_c is the length of the channel, C_{go} is the capacitance of the gate oxide, V_D is the drain voltage and W_c is the width of the channel. $C_{\text{go}} = \epsilon_{\text{go}} / t_{\text{og}}$ where ϵ_{go} is the dielectric constant of the gate oxide and t_{og} is the thickness of the gate oxide. This device shows good electronic performance. High mobility of 32 cm²/V s and an ON/OFF ratio of 10^6 is attained, which is comparatively higher than the previous reported devices fabricated on CVD-grown MoS₂ monolayers.^{22,26,27}

Conclusion

We have demonstrated the optical and electronic properties of a FET device based on MoS₂ flakes grown by CVD. A uniform, highly crystalline, large-area, high-quality monolayer of MoS₂ is achieved by CVD on SiO₂ substrates. Electrodes are fabricated by optical lithography and MoS₂, grown by CVD, is transferred through PC film. AFM, Raman, PL, and back-gated measurements are performed in ambient conditions without any encapsulation of the device. The fabricated device reveals n -type behavior with high field-effect mobility of 32 cm²/V s and high current ON/OFF ratio of 10^6 . Good ohmic contact is achieved when using In as source/drain electrodes. The large-sized, highly crystalline flakes of MoS₂ and the fabricated device showing high field-effect mobility and ON/OFF ratio make them potential candidates for high-performance nanoelectronic and optoelectronic devices.

Conflict of interest The authors declare that they have no conflict of interest.

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