Research article

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Facile integration of MoS₂/SiC photodetector by direct chemical vapor deposition

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Abstract: The MoS₂ photodetector on different substrates stacked via van der Waals force has been explored extensively because of its great potential in optoelectronics. Here, we integrate multilayer MoS₂ on monocrystalline SiC substrate through direct chemical vapor deposition. The MoS₂ film on SiC substrate shows high quality and thermal stability, in which the full width at half-maximum and first-order temperature coefficient for the E_{2g}^1 Raman mode are 4.6 cm⁻¹ and -0.01382 cm⁻¹/K, respectively. The fabricated photodetector exhibits excellent performance in the UV and visible regions, including an extremely low dark current of ~1 nA at a bias of 20 V and a low noise equivalent of 10⁻¹³–10⁻¹⁵ W/Hz^{1/2}. The maximum responsivity of the MoS₂/SiC photodetector is 5.7 A/W with the incident light power of 4.35 μW at 365 nm (UV light). Furthermore, the maximum photoconductive gain, noise equivalent power, and normalized detectivity for the fabricated detector under 365 nm illumination are 79.8, 7.08 × 10⁻¹⁵ W/Hz^{1/2}, and 3.07 × 10¹⁰ Jones, respectively. We thus demonstrate the possibility for integrating high-performance

photodetectors array based on MoS₂/SiC via direct chemical vapor growth.

Keywords: MoS₂; SiC; chemical vapor deposition; photodetector.

1 Introduction

Two-dimensional, layered molybdenum disulfide (MoS₂) has been attracting widespread attention because of its fascinating properties for potential applications in modern electronics and optoelectronics [1, 2]. A monolayer of MoS₂ has been shown to exhibit a direct bandgap of 1.8 eV, while bulk MoS₂ has an indirect bandgap of 1.2 eV [1]. The tunable bandgap of MoS₂ by thickness modulation provides great advantages in optoelectronic devices such as phototransistors and photodetectors [3, 4]. Although MoS₂ transistors based on exfoliated flakes have been demonstrated with high I_{on}/I_{off} ratio and high field-effect mobility, the disadvantages of uncontrollable flake size and random number of layers hinder its performance optimization and reproducibility [5]. Chemical vapor deposition (CVD) has been widely used because of its practical superiority in large-scale integrated electronics [6–9]. For instance, graphene prepared by CVD has been widely applied in ultrafast fiber lasers for soliton study [10–12]. The CVD approach can not only give wafer-scale MoS₂ films with a specified thickness but also enable its direct growth on substrates, which would largely avoid interfacial contamination during the exfoliated MoS₂ transfer process [13]. Moreover, the effective low-dimensional hybrid heterostructure can improve the performance of a device [14, 15]. Nowadays, MoS₂-based hybrid heterojunctions are integrated generally as photodetectors, such as MoS₂/GaN [16–18], MoS₂/AlN [19], MoS₂/graphene [20, 21], and so on [22–24]. Li et al. demonstrated vertically stacked MoS₂/GaN heterojunctions with a strong enhancement of electroluminescence [25]. Liao et al. also reported that heterojunctions of MoS₂/GaN and MoS₂/AlN show good optical adsorption in ultraviolet and visible regions [26].

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These investigations have shown the great potential of two-dimensional/three-dimensional (2D/3D) heterojunction-based devices.

Silicon carbide (SiC) is also a promising candidate for ultraviolet (UV) photodetectors because of its unique properties such as a wide bandgap, high breakdown electric field, and high thermal stability [27–29]. With a wide bandgap of ~3.2 eV, SiC can be used in UV photodetectors, whereas MoS₂ shows a broad spectral response varying from the visible to the near-infrared region depending on its bandgap. Consequently, a photodetector with a wide detectivity range can be realized by integrating MoS₂ with SiC substrates. Lee et al. studied the current-voltage (*I*-*V*) and capacitance-voltage (*C*-*V*) characteristics of heterojunctions between p-type MoS₂ and n-type 4H-SiC [30]. The lattice mismatch (2.90%, $\alpha_{\text{MoS}_2} = 3.162 \text{ \AA}$, $\alpha_{\text{4H-SiC}} = 3.073 \text{ \AA}$) and the difference in thermal expansion coefficients ($\alpha_{\text{MoS}_2} = 1.90 \times 10^{-6} \text{ K}^{-1}$, $\alpha_{\text{4H-SiC}} = 4.47 \times 10^{-6} \text{ K}^{-1}$) between MoS₂ and 4H-SiC may contribute to interfacial strain [31–33]. This has been demonstrated in a number of strain-induced changes in 2D materials [34–37]. Essentially, thermal conductivity is related to phonon-phonon scattering, which is caused by structure buckling [38, 39]. Though mechanically exfoliated MoS₂ on SiC has shown a substrate-enhanced photogating effect [40], the optical-electrical characteristics of MoS₂/SiC photodetectors integrated directly by CVD have not been reported yet.

In this work, we successfully fabricated MoS₂/SiC photodetectors through direct CVD. The quality as well as optical and thermal stability characteristics of multilayer MoS₂ on monocrystalline SiC substrate were characterized via Raman spectroscopy, X-ray photoelectron spectroscopy (XPS), transmission electron microscopy (TEM), energy dispersive spectroscopy (EDS), and absorption spectroscopy. In addition, the photoresponse of MoS₂/SiC photodetectors under UV and visible light illumination was systematically investigated.

2 Experimental section

2.1 Integration of MoS₂ on SiC substrate and preparation of the photodetector

Prior to the growth MoS₂ film, the 4H-SiC substrate went through a precleaning process, which consisted of a degreasing step of 5 min in acetone and 5 min in isopropanol, and a 2-h immersion in Piranha solution (H₂O₂:H₂SO₄=1:3) for the removal of organic residues. Large-area, continuous, multilayer MoS₂ were synthesized on (0001)-oriented 4H-SiC substrates by a CVD system

with two zones. MoO₃ powder was spread uniformly on the side near the Ar source in an alumina crucible, and 4H-SiC substrates were placed at the center of crucible. Sulfur powder was placed upstream in 50 sccm of Ar gas flow. Multilayer MoS₂ film was grown at 750°C for 8 min. Because of the limitation of the CVD chamber diameter, the 4H-SiC substrate was cut into small pieces (1 cm × 1 cm) for CVD growth. Moreover, the MoS₂ film was also grown on a SiO₂/Si substrate as the control sample for comparison, as schematically illustrated in Figure S1. After growing the MoS₂ film, it was transferred to the SiC substrate by a poly(methyl methacrylate) (PMMA)-assisted method. Through a KOH etching process, the MoS₂ film covered by PMMA floated on the top of the solution. Then, a cleaning step in deionized water was performed to remove any possible residual K⁺ on the sample, followed by its transfer on to the SiC substrate. In the end, the PMMA layer was dissolved away by acetone. The pattern of the electrodes was defined using UV lithography, and the electrodes of Ti (5 nm)/Au (50 nm) were formed via e-beam evaporation.

2.2 Characterization

The room-temperature and temperature-dependent Raman spectral measurements were performed using a RENISHAW spectrometer (Renishaw, Wotton-under-Edge, Gloucestershire, United Kingdom) with the excitation wavelength of 514.5 nm, in ambient air. In order to avoid Joule heating of the sample, the laser power was set to 0.25 mW. The band alignment of MoS₂/SiC interface was determined via XPS measurements (Thermo ESCALAB 250Xi, Thermo Fisher Scientific, Waltham, MA, USA). The excitation was through a monochromated Al source ($h\nu = 1486.6 \text{ eV}$) at 150 W. All XPS spectra were calibrated to the C 1s electron peak at 284.8 eV, and the elemental spectra were recorded in steps of 0.05 eV. A UV-visible spectrometer (Shimadzu-2450, Tokyo, Japan) was used to characterize the absorbance of the film in the range 400–800 nm. The electrical characteristics of the MoS₂/SiC photodetector were studied by a Keithley 4200-SCS semiconductor parameter analyzer (Keithley, Beaverton, OR, USA) at room temperature.

3 Results and discussion

Figure 1A shows a schematic of the CVD equipment used to grow MoS₂ thin films on SiC substrates. Powders of S and MoO₃ were the precursors for synthesizing MoS₂ films on SiC, and the sample growth was completed in Ar atmosphere at 750°C. Figure 1B shows the Raman spectra of

Figure 1: Synthesis and characterizations of MoS₂ grown on SiC by direct CVD.

(A) Schematic of MoS₂ grown on monocrystalline SiC by direct CVD under Ar atmosphere, in which MoS₂ was deposited on the Si face of SiC. (B) Raman spectrum of the MoS₂ on SiC. The difference between the Raman peaks E_{2g}^1 and A_{1g} is 23.9 cm⁻¹. The FWHM of the E_{2g}^1 peak is 4.7 cm⁻¹. (C) Cross-sectional TEM image of the MoS₂/SiC sample. (D) Absorbance of MoS₂, SiC, and MoS₂ on SiC. The strongest absorption is at around 440 nm.

multilayer MoS₂ grown on monocrystalline SiC substrate (G-MoS₂). The E_{2g}^1 and A_{1g} optical mode peaks were detected at 380.9 and 404.8 cm⁻¹, which are due to the in-plane vibration of two S atoms with respect to the Mo atom and the out-of-plane vibration of only the S atoms in opposite directions, respectively [41]. Compared to the single-layer MoS₂ [42, 43], there is a blue shift in the E_{2g}^1 peak position of multilayer MoS₂, while the A_{1g} peak shows a red shift. This is due to the sensitive thickness dependence of the E_{2g}^1 and A_{1g} modes [42]. The difference in peak positions of the E_{2g}^1 and A_{1g} modes is about 23.9 cm⁻¹, indicating a quadrilayer MoS₂ film, which is well consistent with a previous report [44]. The full width at half-maximum (FWHM) of the E_{2g}^1 peak, which is related to the crystallinity of MoS₂, is 4.7 cm⁻¹, implying the highly crystalline nature of MoS₂. The MoS₂ film transferred to the SiC substrate (T-MoS₂) was also characterized by its Raman spectrum, shown in Figure S2. Notably, the FWHM of E_{2g}^1 for T-MoS₂ is 5.7 cm⁻¹, which is larger than that of G-MoS₂. The difference in peak positions between E_{2g}^1 and A_{1g} is also about 23.9 cm⁻¹, indicative of a quadrilayer MoS₂ film. The thickness of MoS₂ is about 2.63 nm, and a quadrilayer MoS₂ film was further confirmed in TEM measurements, as shown in Figure 1C, agreeing well with the Raman spectra. The bandgap of a quadrilayer MoS₂ film is 1.4 eV [45], which means that the MoS₂/SiC detector in this work will respond to incident light in the wavelength range below ~886 nm. In addition, no obvious interface layer between MoS₂ and SiC was found. The TEM-EDS images shown in Figure S3 illustrate the interfacial elemental distribution of MoS₂/

SiC, including sulfur, molybdenum, carbon, silicon, and oxygen. The latter could be due to the oxidation of SiC at high temperatures, in which the oxygen could come from MoO₃. In order to identify the optical properties of MoS₂ thin films, the absorbance spectra of MoS₂, SiC, and MoS₂ grown on SiC as a function of incident wavelength were measured. As shown in Figure 1D, the strongest absorbance (A) peak of MoS₂ occurs at ~440 nm. With the MoS₂ film grown on monocrystalline SiC, the MoS₂/SiC heterostructure shows a higher absorbance. Moreover, due to the SiC bandgap of ~3.2 eV [46], there are no obvious absorption peaks above 400 nm for MoS₂/SiC. It is to be noted that A is defined as $\log_{10}(I_0/I_t)$, where I_0 and I_t are the intensities of the incident and transmitted light, respectively. Therefore, according to the Beer-Lambert law, $I_t = I_0 e^{-\alpha t}$ (where t represents the thickness of material and α the absorption coefficient), the absorption coefficient α can be deduced accordingly [47]. When the incident wavelength is about 460 nm (or 660 nm), A is 0.21 (or 0.051) for multilayer MoS₂. Based on the above, the absorption coefficient α in our case is 1.8×10^6 cm⁻¹ (or 4.5×10^5 cm⁻¹). Note that the extra absorption due to the SiC substrate has been removed. Consequently, the high absorption coefficient will raise the material's response to light, resulting in a highly responsive MoS₂ photodetector on the SiC substrate.

Because of the self-heating effect, it is important to examine the material's performance under different temperatures. The temperature-dependent Raman spectrum is used to probe the lattice vibration at different temperatures. The variation of the Raman peak position is related

Figure 2: Temperature dependent Raman spectra for MoS₂ on the SiC.

(A) Raman spectra of multilayer MoS₂ grown on a SiC substrate at different temperatures. (B) Temperature dependence of the peak positions of A_{1g} and E_{2g}¹ Raman modes of MoS₂ on SiC.

to the material's thermal stability. Figure 2A shows the Raman spectra of a multilayer MoS₂ sample grown on SiC substrate at different temperatures in the range 300–500 K in steps of 20 K. As the temperature is increased, the Raman peaks of E_{2g}¹ and A_{1g} show a red shift, which is consistent with the observation from other two-dimensional materials [48, 49]. The temperature dependence of the Raman E_{2g}¹ and A_{1g} peaks was extracted in detail, as shown in Figure 2B, by fitting with the following equation [50]:

$$\omega = \omega_0 + \chi T \quad (1)$$

where ω_0 is the Raman peak position of the E_{2g}¹ (or A_{1g}) mode at 0 K, T is the temperature in kelvin, and χ presents first-order temperature coefficient. By a linear fitting, the χ values of the E_{2g}¹ and A_{1g} Raman modes of multilayer MoS₂ were calculated as -0.01382 and $-0.01214 \text{ cm}^{-1}/\text{K}$, respectively. In contrast with the temperature coefficient (χ) of multilayer MoS₂ grown on glass or SiO₂ substrates [50–52], that of the E_{2g}¹ and A_{1g} Raman modes on SiC substrates is much smaller. This indicates that MoS₂ grown on SiC has better thermal stability. Another interesting observation is that the χ value of the E_{2g}¹ Raman mode shows a larger difference than what was previously reported. It could be due to the lattice mismatch caused by growing the MoS₂ film on SiC substrates, since the E_{2g}¹ Raman mode is sensitive to strain and phonon vibrations; also, the thermal conductivity of MoS₂ can be affected by the tensile strain [34, 53].

The 3D schematic of the structure of MoS₂ photodetectors with Ti/Au electrodes is shown in Figure 3A, where the channel length of the MoS₂ photodetector is 15 μm .

Figure 3B shows the optical microscopy image of multilayer MoS₂/SiC photodetector arrays. The larger-scale MoS₂ film was deposited on monocrystalline SiC substrate by CVD, which completely covered a $1 \times 1 \text{ cm}^2$ SiC substrate. The atomic force microscopy (AFM) images of the electrode and the multilayer MoS₂ film are shown in Figure S4. Figure 3C shows the photoelectric characteristics of the detectors that were constructed by transferring MoS₂ (T-detector) and those of directly grown MoS₂ (D-detector), with an incident light power of $4.35 \mu\text{W}$ at 365 nm. The dark currents of both (T- and D-detector) are about 1 nA. This implies that MoS₂ grown on SiC has the same carrier concentration. Moreover, under the same light surroundings, the photocurrent of the D-detector is significantly larger than that of the T-detector. This photocurrent gap is close to two orders of magnitude at a bias of 20 V. For investigating the influence of the substrate, the band alignment of MoS₂/SiC interface was determined using the Kraut method [54]. Figure S5 shows the high-resolution XPS spectra for the core level energy and valence band maximum (VBM) of three samples, namely multilayer MoS₂ on SiO₂/Si, bulk SiC, and multilayer MoS₂ grown on SiC. The bandgaps of multilayer MoS₂ and SiC are 1.40 eV [45] and 3.25 eV [46], respectively. The conduction and valence band offsets were determined to be 2.09 and 0.24 eV, respectively, with the formation of type-II heterojunctions. The band alignment diagram of MoS₂/SiC is shown in Figure S6. Moreover, the type-II band alignment between MoS₂ and SiC will result in a higher possibility that electrons in SiC will flow toward MoS₂. It will enhance the separation of photogenerated electron-hole pairs at the interface of MoS₂/SiC. In other words,

Figure 3: The structure and I - V characteristics of MoS₂/SiC photodetector.

(A) 3D structure diagram of the MoS₂/SiC photodetector, in which the electrodes is Ti/Au. (B) Optical microscope image of MoS₂/SiC photodetectors arrays. (C) I - V characteristics of detectors constructed by transferring MoS₂ (T-detector) and directly grown MoS₂ (D-detector) under dark and light illumination, respectively. (D) Photocurrent as a function of the incident light power for T- and D-detectors, respectively.

more photogenerated carriers could be accumulated at the interface due to the SiC substrate enhancement, which is called the dual-photogating effect [40]. Figure 3D shows the photocurrent as a function of the incident light power for T- and D-detectors. It is seen that the photocurrent slightly increases as the incident light power is increased and then reaches saturation. This could be explained via the saturation absorption or electron-hole pair recombination [55–57].

Figure 4A shows the dark current and photocurrent of the MoS₂ photodetector under illumination with 365 nm but with different incident light powers. The dark current is not more than 10 nA even at a bias of 20 V. With respect to symmetrical electrodes, the generation of photocurrent could be caused by the change of the photoconductor for MoS₂ through optical doping [58]. The separation of electron-hole pairs adjusts the conductive of channel under the applied electric field. When the photodetector is illuminated by a 365 nm laser of 2.163 μW, the photocurrent is up to 6 μA. The ratio I_{ph}/I_{dark} , where $I_{ph} = I_{tot} - I_{dark}$, is about 3×10^4 . Figure 4B shows the time-dependent photocurrent under continuous and pulsed light illumination. The photocurrent during the ON and OFF states is set at 0.1 Hz photoswitching rate. To avoid instability of the photocurrent, the incident light power is set as 5.647 μW as the photocurrent saturates, where the applied bias is 10 V.

As shown in the inset of Figure 4B, the rise time (τ_r) and fall time (τ_d) of the fabricated photodetector are 0.58 and 0.33 s, respectively. Thus the device exhibits a good stability and repeatability in time-dependent photocurrent.

We also examined the photoresponse and photo-switching behaviors under illumination with 460 and 660 nm, as shown in Figure 4C–F. The photoresponse of the MoS₂ photodetector is summarized in Table S1. For the incident light of 460 and 660 nm, the ratio I_{ph}/I_{dark} is about 380 and 50, respectively. The saturation of photocurrent with increase in incident light power also occurred. When the photodetector is under illumination of 460 nm (or 660 nm) laser with the saturated incidence light power at a bias of 20 V, the current is up to 0.77 μA (or 0.31 μA). The photocurrent under the light illumination at 460 nm (or 660 nm) is far lower than that at 365 nm. Both of τ_r and τ_d show wavelength-dependent behaviors, but τ_d is much more sensitive. For incident light of wavelengths 460 and 660 nm, τ_r (or τ_d) is 0.75 s (or 0.80 s) and 1.08 s (or 1.10 s). The wavelength-dependent behavior could be attributed to the difference in absorbance of MoS₂/SiC under difference wavelengths. On excitation with photon of a particular energy, the photogenerated carriers in SiC will separate. Then, a number of positive charges will accumulate near or at the interface of MoS₂/SiC to effectively produce a vertical electric field, which will promote the

Figure 4: Photoresponse characteristics of MoS₂/SiC photodetector.

Photoresponse (I - V) characteristics of MoS₂/SiC under (A) 365 nm, (C) 460 nm, and (E) 660 nm incident light with different light powers.

Typical time-dependent current curve of the photodetector at bias of 10 V under (B) 365 nm, (D) 460 nm, and (F) 660 nm. The insets show the corresponding rise time and fall time for fabricated MoS₂/SiC photodetectors.

photogeneration of carriers in MoS₂ channel. The SiC substrate can trap the incident photon at the UV and visible spectrum, introducing an extra photoelectric effect, which could result from the wide bandgap and in-gap impurity levels [59–61]. When the energy of the incident photon is greater than the bandgap energy of SiC, band-to-band absorption will occur to provide more carriers, resulting in a higher vertical electric field. It represents the generation of more electron-hole carriers at a given time, that is to say, τ_r (or τ_d) is shorter. The response speed

of the detector is mainly determined by the carrier lifetime, which is related to the density of the surface defect states [62]. Compared with other MoS₂ detectors via the large-area preparation method [16, 63], the present device shows a prolonged photocurrent switching time. This could be due to the defects introduced during the deposition of the MoS₂ film on SiC. A higher Schottky barrier height between MoS₂ and the metal can reduce the photoresponse time by accelerating the separation of electron-hole pairs [64, 65].

Figure 5A–D shows the responsivity R as the function of the incident light power at different incident wavelengths under a bias of 20 V. R is defined by

$$R_i = \frac{I_{\text{tot}} - I_{\text{dark}}}{P_\lambda} \quad (2)$$

where P_λ is the incident light power per unit effective area. As the incident wavelength is 365 nm, the peak of R under 365 nm illumination is 5.7 A/W with 4.351 μW . R and P_λ show a positive correlation when P_λ is less than 4.351 μW . The reverse trend is seen when P_λ is higher than 4.351 μW . The highest R under 460 and 660 nm are 0.51 and 0.224 A/W,

respectively, which are $10\times$ less than that at 365 nm. The ultra-large R can be ascribed to the higher absorbance for the incident light at the lower wavelength. Furthermore, The G - P_λ curves under different incident wavelengths show the same trends as the R - P_λ curves, in which

$$G = \frac{I_{\text{ph}}/e}{\mu P_\lambda / h\nu} \quad (3)$$

where μ is the absorption percentage, e is the elementary charge, h is the Planck constant, and ν is the frequency of the incident light. In addition, G at 365 nm is also much higher than at 460 and 660 nm. It means that higher

Figure 5: Incident power evolution of G , R , NEP and D^* for MoS₂/SiC photodetector.

Photoconductive gain (G) and responsivity (R) of the MoS₂/SiC D-detector as a function of the incident light power for three incident light wavelengths (A) 365, (C) 460, and (E) 660 nm at a bias of 20 V. Noise equivalent power (NEP) and normalized detectivity (D^*) of MoS₂/SiC D-detector as a function of incident light power for three incident light wavelengths (B) 365, (D) 460, and (F) 660 nm at a bias of 20 V.

energy incident photons can generate more electron-hole pairs in the channel. The highest value of G is 79.8 in our devices. When the photodetector is under 460 or 660 nm illumination, the maximum G is 3.8.

A high-performance photodetector has a low noise equivalent power (NEP) and higher D^* [66]. Moreover, NEP represents the level of excitation power that generates a signal equal to the noise under 1 Hz bandwidth. NEP can be calculated by

$$\text{NEP} = \frac{(A_0 \Delta f)^{1/2}}{D^*} \quad (4)$$

where A_0 is the effective area of illumination and Δf is the bandwidth, and D^* is dominated by three factors, namely dark-current-induced shot noise, Johnson-Nyquist noise, and thermal fluctuation (“flicker”) noise. Usually, the dark current is the major contributor. Therefore, D^* can be simplified as

$$D^* = \frac{R_i}{(2eI_{\text{dark}})^{1/2}} \quad (5)$$

All data are from measurements at a bias of 20 V, as shown in Figure 5B–F. Among the three wavelengths, the lowest NEP is 7.08×10^{-15} W/Hz^{1/2} and the highest D^* is 3.07×10^{10} Jones. Comparison of the photoresponses of MoS₂ heterojunction detectors is summarized in Table S2 [67–72]. For improving the photocurrent switching of the fabricated MoS₂/SiC photodetector, proper optimization of the device passivation and the metal electrode should be done.

4 Conclusion

In summary, we have demonstrated a high-performance MoS₂/SiC photodetector by using a direct CVD method. The high quality and thermal stability of MoS₂ thin film were characterized by Raman measurement. The MoS₂ D-detector showed a higher photocurrent than the MoS₂ T-detector and a low dark current of 1 nA under 365 nm laser illumination. Under UV and visible light illumination, the MoS₂ D-detector showed excellent performance with a low NEP of 10^{-13} – 10^{-15} W/Hz^{1/2} and a lower typical response time of 0.33–1.10 s. Its responsivity could reach 5.7 A/W under UV light illumination. Our one-step, integrated MoS₂/SiC photodetectors can provide insights into the possibility for integrating high-performance photodetector arrays.

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