Research article

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High-performance monolayer MoS₂ photodetector enabled by oxide stress liner using scalable chemical vapor growth method

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Abstract: MoS₂, as a typical representative of two-dimensional semiconductors, has been explored extensively in applications of optoelectronic devices because of its adjustable bandgap. However, to date, the performance of the fabricated photodetectors has been very sensitive to the surrounding environment owing to the large surface-to-volume ratio. In this work, we report on large-scale, high-performance monolayer MoS₂ photodetectors covered with a 3-nm Al₂O₃ layer grown by atomic layer deposition. In comparison with the device without the Al₂O₃ stress liner, both the photocurrent and responsivity are improved by over 10 times under 460-nm light illumination, which is due to the tensile strain induced by the Al₂O₃ layer. Further

characterization demonstrated state-of-the-art performance of the device with a responsivity of 16.103 A W⁻¹, gain of 191.80, NEP of 7.96×10^{-15} W Hz^{-1/2}, and detectivity of 2.73×10^{10} Jones. Meanwhile, the response rise time of the photodetector also reduced greatly because of the increased electron mobility and reduced surface defects due to the Al₂O₃ stress liner. Our results demonstrate the potential application of large-scale strained monolayer MoS₂ photodetectors in next-generation imaging systems.

Keywords: monolayer MoS₂; Al₂O₃; stress liner; photodetector.

1 Introduction

Ever since the discovery of graphene in 2004, two-dimensional (2D) materials have become a hot topic of research owing to their special properties such as the absence of surface dangling bonds, large surface-to-volume ratio, energy bandgap tunable with the number of layers, as well as adjustable chemical and physical properties [1–5]. Among them, semiconducting transition-metal dichalcogenides (TMDCs) have been considered as promising materials for future and emerging electronic, optoelectronic, and biological devices [6, 7]. MoS₂, a typical representative of TMDCs, possesses a bandgap ranging from 1.80 eV (monolayer) to 1.20 eV (bulk), high mobility, high light absorptivity, and thermal stability (~1100°C), which facilitate its applications in field-effect transistors (FETs), photodetectors, and sensors [1, 8–11]. For photodetectors, monolayer or multilayer MoS₂-based phototransistors have made significant progress. The exfoliated monolayer MoS₂ phototransistor shows a responsivity of up to ~880 A W⁻¹ [12], while the detectivity and responsivity of a multilayer MoS₂ photodetector reach 10¹² Jones and 2570 A W⁻¹ [13], respectively. However, it is still a great challenge to achieve large-area, high-quality monolayer MoS₂ films. Chemical vapor deposition (CVD) as a promising method has been widely employed. Yore et al. have

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successfully grown large-area monolayer MoS₂ films on sapphire substrates and made them into photodetectors with a responsivity of 0.8 mA W⁻¹ [14]. Obviously, such low responsivity is unable to meet the requirements for detection capabilities. Thus, finding ways to improve the responsivity of MoS₂-based photodetectors is an urgent need. In the literature, a common method to improve the performance of photodetectors is to form a heterojunction with graphene [15–20]. Fazio et al. prepared a graphene/MoS₂ heterojunction photodetector on a flexible substrate, giving a responsivity of up to 45.5 A W⁻¹ [21]. In addition, the MoS₂/TiO₂ p-n heterojunction and the n-doped MoS₂ photodetector achieved responsivity values of 35.9 [22] and 99.9 A W⁻¹ [23], respectively. Another efficient way is to protect the MoS₂ from the environment by covering it with Al₂O₃, HfO₂, or BN layers [1, 24–26]. Over 20-fold enhancement of photoresponsivity of multilayer MoS₂ transistors using a high-work-function MoO_x overlayer was also demonstrated [27]. Highly stable and high-performance monolayer/bilayer MoS₂ photodetectors covered with atomic layer deposited (ALD) HfO₂ exhibited zero hysteresis and low device resistance [28]. FETs fabricated on exfoliated MoS₂ flakes capped with a 15-nm Al₂O₃ film achieved an effective carrier mobility of 700 cm² V⁻¹ s⁻¹ at room temperature [24], which is primarily attributed to the screening effect on charged-impurity scattering. In addition, Yu et al. studied the interface effect between Al₂O₃ and monolayer MoS₂ by first-principles calculation, which indicated that the 3-nm Al₂O₃ can introduce about 0.41% tensile strain on monolayer MoS₂ [29]. Large changes in the electrical properties of monolayer MoS₂ caused by doping and strain also affect its application in nanoelectronics [30]. The electron mobility of the monolayer MoS₂ effectively increases by over 10 times by the biaxial strain effect [31]. The strain in monolayer MoS₂ can be detected by Raman spectroscopy, where the E_{2g}¹ mode is sensitive to strain [32]. Until now, most of the reported results are based on exfoliated MoS₂ flakes instead of grown films, and systematic investigations of the effects of strain on the device level have not been carried out. Therefore, it is worthwhile investing some effort to explore this subject.

We have successfully grown high-quality, large area, and continuous monolayer MoS₂ on sapphire substrates using CVD, and deposited a 3-nm-thick Al₂O₃ stress liner grown by ALD to isolate the MoS₂ layer from the environment. X-ray photoelectron spectroscopy (XPS), transmission electron microscopy (TEM), Raman spectroscopy, and absorption techniques were carried out to characterize the material. Then MoS₂-on-sapphire photodetectors with and without the Al₂O₃ stress liner were fabricated using typical semiconductor fabrication processes. After applying the

Al₂O₃ stress liner, not only the photocurrent and photoresponsivity of the photodetector increased profoundly, but also the response time reduced substantially.

2 Experimental section

2.1 Materials

Acetone (CH₃COCH₃, 99.5%), isopropyl alcohol (C₃H₈O, 99.5%), and alcohol (C₂H₆O, 99.7%) were purchased from Aladdin Co., Ltd (Shenzhen, China). Molybdenum trioxide powder (MoO₃, 99.95%) and sulfur powder were purchased from Aladdin Co., Ltd. Trimethylaluminum (TMAI, 99.9999%) was purchased from Fornano Co., Ltd (Suzhou, China). Titanium (Ti, 99.999%) and gold (Au, 99.999%) were purchased from Tim Co., Ltd (Beijing, China). Double-side-polished sapphire substrates (430 μm thickness) with (0001) orientation were purchased from Xiamen Zhonghe Optoelectronics Co., Ltd (Xiamen, China). The photoresist (AZ5214) and developer (ZX238) were purchased from AZ Co., Ltd (Shanghai, China). Deionized water (DI water) was prepared in the laboratory.

2.2 Preparation of the MoS₂ on sapphire, Al₂O₃, and photodetectors

Large-area, continuous monolayer MoS₂ were grown on (0001)-oriented sapphire substrates by a CVD system with two zones. Prior to the growth, the substrates were first cleaned using acetone, isopropanol, and DI water. After the treatment, they were placed face down on a crucible containing MoO₃ and loaded into a heating furnace with a quartz tube of 100 mm outer diameter. Ultrahigh-purity Ar gas was employed as the carrier gas, and the growth pressure was kept at atmospheric. Another crucible containing 0.8 g sulfur powder was located in the adjacent zone, upstream from the sapphire substrates. Before heating, pure Ar gas with a flow rate of 100 sccm was used to purge the tube in order to drive away other gases (e.g. O₂, CO₂ etc.), and it was then maintained at a flow rate of 50 sccm during the growth. The MoO₃ powder was heated up to 750°C over 120 min, while the sulfur powder was first maintained at room temperature for 70 min and then heated to 200°C in 50 min. Subsequently, the growth temperature was kept at 750°C for 15 min and then naturally cooled down to room temperature. Ti(5 nm)/Au(50 nm) was selected as the electrode, which was deposited on monolayer MoS₂ by the UV lithography and e-beam evaporation and lift-off processes. Then, Al₂O₃ oxide was

deposited on MoS₂ by an ALD system, using TMAI and H₂O. TMAI and H₂O produced sufficient vapor pressure at 300°C, and pure nitrogen with a flow rate of 50 sccm was injected into the reaction chamber. The flow rate of pure nitrogen in the reaction chamber was 200 sccm. After the temperature of the reaction chamber reached 300°C for 10 min, the reaction was carried out, during which the temperature was maintained at 300°C throughout the process. In order to deposit 3 nm Al₂O₃ on the surface of MoS₂, the deposition period was selected as 16 cycles.

2.3 Characterization and measurement

The room-temperature and temperature-dependent Raman spectra were recorded using a confocal Renishaw system with a wavelength of 514.5 nm and a power of 0.25 mW for excitation and 2400 grooves/mm gratings for high resolution. XPS spectra were measured using a VG Escalab 220i-XL system with a constant pass energy of 20 eV and a monochromatic AlK_α (1486.6 eV) X-ray source. The C 1s peak (284.8 eV) was used to correct the core-level binding energy in order to eliminate the differential charging effect on the sample surface. Absorbance spectra were measured using a Shimadzu-2450 UV-visible spectrometer in the spectral range 400–800 nm. The *I*-*V* characteristics and responsivity measurements of the fabricated photodetectors were carried out by a Keithley 4200-SCS semiconductor analyzer.

2.4 Theoretical simulation

First-principles calculations involved in this work were carried out using Material Studio, as implemented in the Cambridge Sequential Total Energy Package (CASTEP) based on density functional theory (DFT) [33]. The super-soft pseudopotential was used to describe the interaction between ions and the valence electrons. The exchange correlation used was the generalized gradient approximation (GGA) in the form of Perdew-Burke-Ernzerhof (PBE) [34], together with the projector augmented wave potential (PAW) to treat ion-electron interactions [35]. The maximum Hellmann-Feynman force was 0.05 eV Å⁻¹, which is sufficient to obtain relaxed structures [34]. The electronic wave functions were expanded in a plane wave basis set with an energy cut-off of 650 eV to ensure convergence, and a 5×5×1 Monkhorste-Pack mesh was used in the Brillouin zone sampling [36]. The convergence criteria for the geometric optimization of the system were as follows: total energy changes during the optimization finally converged

to less than 10⁻⁶ eV/atom, maximum force on each atom in the crystal was less than 0.03 eV nm⁻¹, the maximum stress in the crystal was less than 0.05 GPa, and the maximum displacement of the atom was less than 10⁻⁴ nm.

3 Results and discussion

Double-polished sapphire with surface orientation (0001) was used as the substrate for the growth of monolayer MoS₂ films. In order to characterize the film thickness and uniformity, room-temperature Raman spectroscopy and TEM were carried out. The Raman spectra of the sample measured at four different positions (marked by 1, 2, 3, and 4 shown in Figure 1A) confirmed that the two main peaks at 384.3 and 403.4 cm⁻¹ belonged to the *E*_{2g}¹ mode corresponding to the in-plane atom vibration and the *A*_{1g} mode corresponding to out-of-plane vibration, respectively. The separation between two peaks is about 19.0 cm⁻¹, indicating that there is only one layer, which is also proved by the cross-sectional TEM image with the thickness of ~0.7 nm in Figure 1B. The TEM-EDS (energy dispersive spectroscopy) image shown in Figure S1A–E illustrates the distribution of Mo, S, Al, and O elements, respectively. The full width at half-maximum (FWHM) of the *E*_{2g}¹ Raman mode is 4.99 cm⁻¹ and that of the *A*_{1g} mode is 8.26 cm⁻¹. The Raman spectral bands at four different positions have almost the same peak position and FWHM, which indicates that the grown sample has excellent uniformity. Figure S2 displays the XPS spectrum showing the core energy levels of Mo and S atoms. The binding energy of Mo 3d_{3/2}, Mo 3d_{5/2}, and S 2s peaks are determined to be 233.23, 230.13, and 227.23 eV, respectively, which is consistent with the reported values [37]. In addition, no peak related to the Mo-O bond was found near 235.6 eV, implying that no byproduct MoO_x was synthesized during the growth of MoS₂. The atomic ratio of S to Mo is calculated to be about 1.98, which is close to stoichiometric MoS₂, and the S 2s peak observed at 227.23 eV is the characteristic of stoichiometric MoS₂ [38, 39]. The TEM characterization was also carried out on the stress liner sample where the monolayer MoS₂ was covered by Al₂O₃. The TEM result of the stress liner sample demonstrated that the thickness of Al₂O₃ above MoS₂ was about 3 nm, as displayed in Figure S3.

In order to further understand the effects of the Al₂O₃ stress liner on the thermal properties of monolayer MoS₂, temperature-dependent Raman spectra were recorded to investigate the lattice vibration properties, which are important for the related devices. Figure S4A,B shows the Raman spectra of both the control sample and the

Figure 1: Material characterization of MoS₂ with and without oxide stress liner.

(A) Raman spectra of a monolayer MoS₂ sample grown on a 1 cm × 1 cm sapphire substrate from the four locations (indicated by the 1, 2, 3, and 4 in the inset). The distance between the E_{2g}¹ and A_{1g} modes is 19 cm⁻¹. (B) Cross-sectional transmission electron microscopy (TEM) image of the monolayer MoS₂/sapphire sample. (C) E_{2g}¹ and A_{1g} Raman modes of control and stress liner samples as a function of temperature including a linear fit. (D) Raman spectra of the control and stress liner samples at room temperature.

sample with the Al₂O₃ stress liner sample as a function of the temperature, which was varied from 300 to 500 K in steps of 20 K. Meanwhile, Figure 1C shows the peak positions of the E_{2g}¹ and A_{1g} Raman modes as a function of temperature for monolayer MoS₂ with and without the stress liner. Obviously, both E_{2g}¹ and A_{1g} Raman phonon modes shift toward the low-frequency side (red shift) with increasing temperature, similar to those of other 2D materials [40–43]. The results can be fitted by the linear equation [44]

$$\omega = \omega_0 + \chi_T \Delta T \quad (1)$$

where ω_0 is the mode frequency measured at room temperature (300 K), χ_T is the first-order temperature coefficient,

and ΔT is the temperature difference relative to 300 K. The measured χ_T values for E_{2g}¹ and A_{1g} in the stress liner sample are about -0.01303 and -0.00927 cm⁻¹ K⁻¹, respectively, which (the absolute value) are much smaller than the corresponding values (-0.01759 and -0.00999 cm⁻¹ K⁻¹, respectively) of the control sample, indicating the better thermal stability of the former, similar to the literature results which show that the Al₂O₃ capping layer significantly improves the thermal stability of black phosphorus [45, 46]. Ding et al. have demonstrated that a tensile strain can soften the phonon vibrations and reduce the thermal conductivity of MoS₂ [47]. Therefore, the tensile strain introduced by the Al₂O₃ capping layer in the stress liner sample plays a dominant role in the improvement of the former's thermal stability.

In contrast, the Raman spectra of the control and stress liner samples are shown in Figure 1D. An obvious red shift of $\omega = -1.22 \text{ cm}^{-1}$ in the stress liner sample can be seen on the E_{2g}^1 peak, while there is no significant change in the A_{1g} and the sapphire-related peaks. The primary cause of this red shift is the effect of tensile strain in the MoS₂ layer introduced by the 3-nm Al₂O₃ stress liner because the E_{2g}^1 mode is a strain-sensitive mode [48]. The strain ε in the stress liner sample is calculated to be 0.58% according to the formula [49] $\varepsilon = \omega / (-2.1 \text{ cm}^{-1}/\%)$, which is close to the first-principles simulation results on the strain effect of Al₂O₃/monolayer MoS₂ interface reported by Yu et al. [29]. Therefore, the strain σ in the stress liner sample can be estimated as 1.566 GPa using the formula $E_{\text{Young}} = \sigma / \varepsilon$, where E_{Young} is Young's modulus taken as 270 GPa [50].

To simulate the strain effect of Al₂O₃ on monolayer MoS₂ in a photodetector, Figure 2A shows the stress distribution of a monolayer MoS₂ photodetector with the 3-nm Al₂O₃ stress liner. The MoS₂ layer in contact with Al₂O₃ is

stretched, whereas that in contact with the Ti/Au is compressed. From the stress distribution of the MoS₂ layer along the horizontal direction, shown in Figure 2B, the tensile stress is higher near the intersection of the monolayer MoS₂, Al₂O₃, and Ti/Au (located at 485 nm) than at other locations. The tensile strain produced by the oxide has a significant effect on the electronic structure of monolayer MoS₂. Figure S5A–E shows, respectively, the first-principles calculation results of the band structures with the tensile strain ranging from 0% to 2% for monolayer MoS₂. The down-shift and up-shift can be observed at the K and Γ points of the reciprocal space of the conduction and valence bands, respectively, with increase in the tensile strain. As a reference, the monolayer MoS₂ without applied strain shows a bandgap of 1.73 eV, which is slightly less than the experimental value of 1.8 eV [51]; this is because the first-principles calculation has the disadvantage of underestimating the bandgap. The bandgap of monolayer MoS₂ has decreased with tensile strain,

Figure 2: Stress analysis of MoS₂ with and without stress liner.

(A) 2D stress mapping within the monolayer MoS₂ photodetector with the Al₂O₃ stress liner. (B) Horizontal stress distribution within the monolayer MoS₂ layer. (C) Tensile strain enables significant reduction of the effective mass of monolayer MoS₂. (D) Calculated absorption coefficient as a function of wavelength under 0–2% tensile strain. (E) Measured absorbance A of control and stress liner samples as a function of the incident wavelength. There are three excitonic absorption peaks (A, B, and C) between 400 and 700 nm in the MoS₂ absorbance curve. The overall absorbance value of the sample with Al₂O₃ is higher than that without it.

changing it from direct bandgap to indirect bandgap, which is consistent with the results reported by Sun and Lin [52]. In addition, the effective mass of the lowest conduction band that the electrons occupy can be calculated through the relation

$$m^* = \hbar^2 / \left(\frac{\partial^2 E}{\partial^2 k} \right) \quad (2)$$

where $\hbar$ is the Planck constant. As shown in Figure 2C, compared with the unstrained monolayer MoS₂, in the strained layer the electron effective mass of MoS₂ is reduced due to the introduction of the tensile strain, which is beneficial for enhancing the electron mobility of MoS₂. Simultaneously, Figure 2D shows that the simulated absorption coefficient of monolayer MoS₂ varies with tensile strain in the wavelength range 440–500 nm. The increase in absorption coefficient with the increase in the tensile strain implies that the tensile strain existing in the MoS₂ layer can effectively enhance the absorption of light.

On the basis of the simulation, the absorbance of the control and stress liner samples was also measured in the experiment. Figure 2E shows the absorbance curves of both samples, which were obtained by subtracting the absorbance of the blank sapphire substrate and plotting it as a function of the incident wavelength. In the absorbance curve of pure MoS₂, two obvious excitonic absorption peaks, labeled as A and B, corresponding to band edges between 600 and 700 nm exist, which originate from the K point of the Brillouin zone. Their energy difference is due to the spin-orbital splitting of the valence band [53]. In addition to excitons associated with the band edges, there is another excitation absorption peak C at 440 nm in Figure 2E. This peak is due to the parallel band or the so-called band nesting effect in the density of states, which produces a strong optical response even though the excitation energy is far beyond the bandgap [54]. Because the “absorbance A ” is defined as $A = \log_{10}(I_0/I_t)$, where I_t is the intensity of the transmitted light and I_0 is the intensity of the incident light, the absorption rate can be obtained by $(I_0 - I_t)/I_0 = 1 - 1/10^A$. In this work, 460 nm was chosen as the wavelength to study the photoelectric performance of the photodetectors. Therefore, the absorption rate of the control sample is determined to be 17.09% at 460 nm incident wavelength, taking $A_{460\text{nm}} = 0.0814$ into account. For the stress liner sample, the absorption rate is 22.83% with $A_{460\text{nm}} = 0.1125$. The absorption ratio increased significantly since Al₂O₃ was deposited onto the MoS₂. According to the Beer-Lambert law

$$I_t = I_0 e^{-\alpha t} \quad (3)$$

where t is the thickness of MoS₂, the absorption coefficient α can be calculated by the following formula:

$$\alpha = \ln(10)A/t \quad (4)$$

Thus, the calculated value of absorption coefficient for the control sample is $3.67 \times 10^6 \text{ cm}^{-1}$ at 460 nm. The corresponding value increases to $6.20 \times 10^6 \text{ cm}^{-1}$ for the stress liner sample, which is due to the effect of the tensile stress in monolayer MoS₂ [55]. Higher absorption of the stress liner sample induced by the Al₂O₃ stress liner can directly result in better performance of the photodetector, especially due to the increased photocurrent.

Transparent arrays of monolayer MoS₂ photodetectors were fabricated with the size $1 \text{ cm} \times 1 \text{ cm}$. Figure 3A shows a 3D schematic view of monolayer MoS₂ photodetectors with the Al₂O₃ stress liner, where a 3-nm Al₂O₃ capping layer was grown using ALD. For the contacts, (5 nm)Ti/(50 nm)Au was deposited as the electrode, which may not be optimal. The contact can be formed by a low-work-function metal such as Sc, which can result in high carrier injection and low contact resistance, since the Fermi level can be pinned close to the conduction band of MoS₂ [24]. Figure 3B shows the current (I) as a function of voltage (V) for both devices plotted under incident light of 460 nm wavelength (1.964 μW) with a laser spot diameter of 1.5 cm. For the control device, the value of the current is 7.1 nA in dark condition at 20 V, suggesting that the monolayer MoS₂ film is undoped and that the Fermi level resides in the bandgap [56]. Meanwhile, the current under illumination is as high as $\sim 1.384 \mu\text{A}$ at 460 nm under an applied voltage of 20 V. When the sample is impinged by a laser, this kind of the observed conductive behavior is due to the generation of photocarriers due to the doping, a process known as optical doping [57]. Under the same condition, the dark current of the device with Al₂O₃ stress liner device increased by two orders of magnitude. The increase in dark current is due to the large tensile strain of MoS₂ near the electrode or to the strain-induced narrowing of the bandgap, as shown in the Figure S5. Similar to the 460 nm in this work, a 660 nm light source was used to perform the same measurement on the photodetector shown in Figure S6. Performance improvement due to the Al₂O₃ stress liner is also seen when the monolayer MoS₂ photodetector was operated under a light source of 660 nm, with the maximum measured photocurrent of 6.387 μA .

The laser-power-dependent I - V curves for the control and stress liner devices under 460 nm illumination are shown in Figure S7A,B. In order to quantify the

Figure 3: IV characteristics of monolayer MoS₂ photodetectors with and without stress liner.

(A) 3D schematic view of monolayer MoS₂ photodetectors with the stress liner. (B) Dark current and light current I as a function of voltage V for the control device and the device with Al₂O₃ stress liner.

photosensitive performance of the photodetector, the photocurrent I_{ph} is extracted by the equation

$$I_{ph} = |I_{light}| - |I_{dark}| \quad (5)$$

where I_{dark} and I_{light} are the current in dark condition and under illumination, respectively. The extracted photocurrent I_{ph} is plotted as a function of the effective power with 460 nm wavelength at a voltage of 20 V in Figure 4A. The curve follows a power law $I_{ph} \propto P^\alpha$, where P is the laser power. For the control device, a linear behavior with $\alpha = 2.31$ can be deduced in the low incident power range of $P_{in} < 1.580 \mu W$, and I_{ph} increases rapidly with the increase

of the power in this range. However, the upward trend of photocurrent is slowed down with $\alpha = 1.24$ in the range $P_{in} > 1.580 \mu W$. There are several possible reasons for this slowdown: (i) absorption reaches saturation [58–60]; (ii) photo-generated carriers are recombined because of existing defects and charge impurities [61]; (iii) recombination of electrons and holes [62]; (iv) exciton-exciton annihilation [63, 64]; and (v) carrier-carrier chattering [59]. Similar behavior for MoS₂ and other semiconductor-based photodetectors has been reported recently [58, 64–66]. In comparison with that of the control device, the “saturation point” is found at a lower power level $P_{in} < 1.454 \mu W$ for the device with the stress liner.

Figure 4: Device characterization under different incident power of monolayer MoS₂ photodetectors with and without stress liner.

(A) Photocurrent with various incident powers measured for a fixed voltage of 20 V at a wavelength 460 nm. The dashed lines indicate the power law ($I_{ph} \propto P^\alpha$) fitting by different α . (B) Performance of control device and the stress liner device as a function of incident power (P) under a fixed voltage (V) of 20 V at 460 nm wavelength. (B) Responsivity (R) and photoconductive gain (G). (C) Noise-equivalent power (NEP) and normalized detectivity (D^*).

The photoresponsivity R is calculated by the expression

$$R = \frac{I_{\text{ph}}}{P_{\text{in}}} \quad (6)$$

where P_{in} is the effective power of the incident light normalized by the equation

$$P_{\text{in}} = \frac{P_0}{\text{power density} \times \text{active area}} \quad (7)$$

where P_0 is the laser output power. To further evaluate the performance of our photodetectors, photoconductive gain G , indicating the number of carriers of the photocurrent that can be generated by one absorbed photon, is also calculated by the equation

$$G = \frac{I_{\text{ph}}}{P_{\text{abs}}} \frac{h\nu}{q} \quad (8)$$

where P_{abs} is the absorbed power, which is defined as $P_{\text{abs}} = \mu P_{\text{in}}$, with μ absorption percentage calculated above and P_{in} the incident power, h is Planck's constant, ν is the frequency of the incident laser, and q is the elementary charge. The calculated photoresponsivity R and photoconductive gain G of both devices are plotted as a function of the effective power at 460 nm wavelength under a bias voltage of 20 V in Figure 4B. For the control device, both R and G show an increasing trend as the irradiation power increases, while for the stress liner device, an increasing trend is seen first below 1.454 μW , and then a decreasing trend above this value. The extracted highest achievable responsivity value in stress liner device is 16.103 A W⁻¹ (1.454 μW laser power), which is more than 16 times higher than that of the control device, which is due to the increased absorption capacity of monolayer MoS₂ with the Al₂O₃ stress liner. In addition, the corresponding photoconductive gain G also increases from 14.20 to 191.80, indicating that the Al₂O₃ stress liner successfully protects the MoS₂ layer from environmental contamination, such as from O₂, H₂O molecules, which in turn reduces the nonradiative recombination ratio [67–69]. In order to further understand the detection limit and signal-to-noise ratio of the photodetectors, the noise equivalent power (NEP) and normalized detectivity (D^*) are shown in Figure 4C. NEP is estimated by the equation

$$\text{NEP} = \frac{\sqrt{2qI_{\text{dark}}\Delta f}}{R} \quad (9)$$

where I_{dark} is the current in the absence of light, R is the responsivity, and f is the bandwidth (here we use $\Delta f = 1$). A photodetector with stronger ability to detect weak signals has a smaller NEP. D^* is defined by

$$D^* = \frac{\sqrt{A}}{\text{NEP}} \quad (10)$$

where A is the device active area. In comparison, the lowest NEP = 7.96 $\times 10^{-15}$ W Hz^{1/2} and highest $D^* = 2.73 \times 10^{10}$ at 460 nm are obtained in the stress liner device, which are one order of magnitude lower and higher, respectively, than the corresponding values of the control device.

In order to understand the reproducibility and speed of the control and stress liner devices, we also measured the temporal response of the photodetectors under a fixed voltage 20 V at 460 nm at ambient air environment. Light with power 1.964 μW is switched on/off alternately and the time with/without illumination is set to 40 s. As shown in Figure 5A,B, both devices show good repeatability and stability without degradation of the photoresponse during the cyclic test. In addition, the corresponding rise time is taken as time to increase from 0% to 90% of the maximum photocurrent and the fall time is taken as that to decrease from 100% to 0% of maximum photocurrent, which are displayed in the inset. The decreasing maximum photocurrent per cycle shown in Figure 5B could be caused by the recombination of electrons and holes due to the defects within the Al₂O₃ layer. The rise time of the control device is 19.0 s, as shown in Figure 5A. In comparison, the corresponding values of the stress liner device is reduced to 12.9 s, as shown in Figure 5B, which is mainly due to the improvement of electron mobility induced by the Al₂O₃ stress liner, but the fall time shows no significant change [70, 71]. Overall, the performance of monolayer MoS₂-based photodetector in this work can be further enhanced by integrating a ferroelectric film or employing a few MoS₂ layers [13, 72].

4 Conclusion

In summary, we successfully fabricated high-quality large-area monolayer layer MoS₂ on transparent sapphire substrates, which was confirmed by Raman, XPS, and TEM characterizations. Temperature-dependent Raman spectra showed that the thermal stability of monolayer layer MoS₂ was improved because of the Al₂O₃ stress liner. A tensile strain of 0.58% was introduced by depositing 3 nm Al₂O₃ with ALD. First-principles calculation based on DFT and the measured absorbance verified the effect

Figure 5: On/off characteristics of monolayer MoS₂ photodetectors with and without stress liner.

Photocurrent as a function of time under alternative darkness and illumination. Photocurrent versus time curve of (A) control device and (B) the stress liner device illuminated by 460 nm light source with the incident power of 1.964 μW at 20 V. The corresponding rise time (from 0% to 90% of maximum photocurrent) and fall time (from 100% to 0% of maximum photocurrent) are shown in the inset.

of the strain introduced by Al₂O₃ on the electron effective mass and light absorption. In comparison with that of the control device, the photocurrent of the stress liner device improved by 20 times and the responsivity by 15 times under illumination of light at 460 nm. A state-of-the-art performance was demonstrated in our stress liner photodetector, leading to a responsivity of 16.103 A W⁻¹, gain of 191.80, NEP of 7.96 × 10⁻¹⁵ W Hz^{-1/2}, and detectivity of 2.73 × 10¹⁰ Jones. The rise time of the photodetector was also shortened because of the increased electron mobility introduced by the Al₂O₃ stress liner. This work demonstrates the potential application of large-scale, strained monolayer MoS₂ photodetectors in next-generation imaging systems.

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