

# Fully Suspended MoS<sub>2</sub> Photodetectors toward High Response Speed and Stable Responsivity

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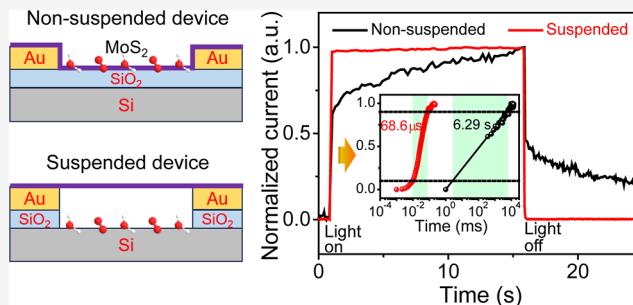
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**ABSTRACT:** High photoresponsivity has been achieved in photodetectors based on two-dimensional (2D) semiconductors, but they usually suffer from low response speed and severe responsivity decay with higher light intensity due to substrate effects. In this work, photodetectors with high response speed and stable responsivity are prepared based on fully suspended MoS<sub>2</sub>. Due to the complete separation of the suspended MoS<sub>2</sub> channel and the Si substrate underneath, the photogating effect is effectively suppressed. Then the photocurrent rise and fall times are measured as 68.6 and 100.0  $\mu$ s by transient photocurrent measurement, respectively, which are about 10<sup>5</sup> times faster than the non-suspended devices. Moreover, they exhibit stable responsivity with time within a large range of light intensity. A detailed analysis of the photoresponse mechanisms is performed by comparing the photoresponses from devices with different structures. The results provide a fundamental understanding of photoresponse mechanisms and guide the design of high-performance 2D photodetectors.

**KEYWORDS:** suspended photodetector, 2D phototransistors, response speed, MoS<sub>2</sub>, transient response, photoresponse mechanism, interface effect



## INTRODUCTION

During the past decade, two-dimensional (2D) semiconducting transition metal dichalcogenides (TMDCs) have been extensively investigated and seen as potential candidates for next-generation electronic and optoelectronic devices due to their unique optical and electrical properties,<sup>1–3</sup> such as atomic-scale thickness, high carrier mobility, tunable bandgap, as well as wafer-scale integration capability.<sup>4,5</sup> Among the various TMDCs, 2D molybdenum disulfide (MoS<sub>2</sub>) attracts lots of attention for its thickness-dependent bandgap, relatively high electron mobility over 200 cm<sup>2</sup>/(V S), and high optical absorption.<sup>6</sup> Phototransistors can be fabricated by simply constructing 2D transistors employing monolayer or multilayer MoS<sub>2</sub> as channel material under light irradiation.<sup>7–9</sup> As reported, phototransistors based on pure MoS<sub>2</sub> channel can achieve ultrahigh responsivity of 880 (monolayer) and 4.3  $\times$  10<sup>3</sup> A/W (multilayer).<sup>10</sup> Moreover, van der Waals heterostructures can further improve the responsivity of 2D phototransistors.<sup>11–13</sup> The responsivity of phototransistors based on multilayer MoS<sub>2</sub> can be even improved to 6.0  $\times$  10<sup>3</sup> A/W by building van der Waals heterostructures between zero-dimensional (0D) silicon quantum dots and 2D MoS<sub>2</sub>.<sup>14</sup>

Although MoS<sub>2</sub>-based phototransistors have achieved excellent responsivity and detectivity, there are still some problems to be solved. One is the low response speed to light. MoS<sub>2</sub>-based phototransistors usually take several seconds to

achieve a saturated current under light illumination,<sup>2,7,15–17</sup> which is far from the practical need. Although photogating effect due to the trap states can enhance the photoresponsivity, it also limits the speed.<sup>18,19</sup> While some techniques such as h-BN encapsulation have been explored to improve the interface, the trap effect from the underlying substrate has not been fully eliminated.<sup>20–22</sup> Another problem is the severe responsivity decay with the increasing incident light intensity,<sup>2,7</sup> which is due to the complex carrier generation, trapping, and recombination processes within the 2D semiconductors.<sup>18</sup>

To address these problems, in this work, we design a new photodetector with a fully suspended MoS<sub>2</sub> channel to separate the MoS<sub>2</sub> channel and Si back gate. The suspended structure effectively suppresses the trap-state-induced photogating effect and simplifies the dynamic process of carrier transport. As a result, the photocurrent rise time and fall time are measured as 68.6 and 100.0  $\mu$ s by transient photocurrent measurement, respectively, which are about 10<sup>5</sup> times faster than the nonsuspended phototransistors. Moreover, the responsivity of

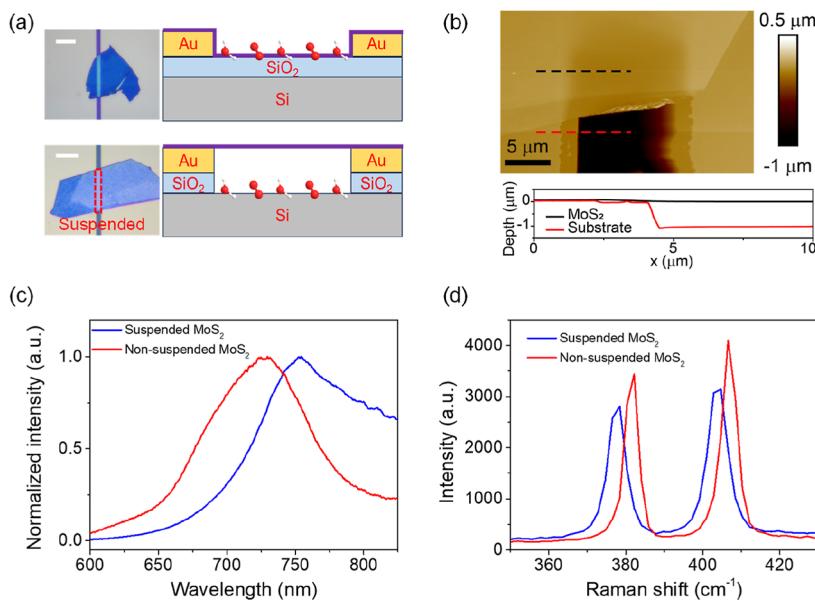
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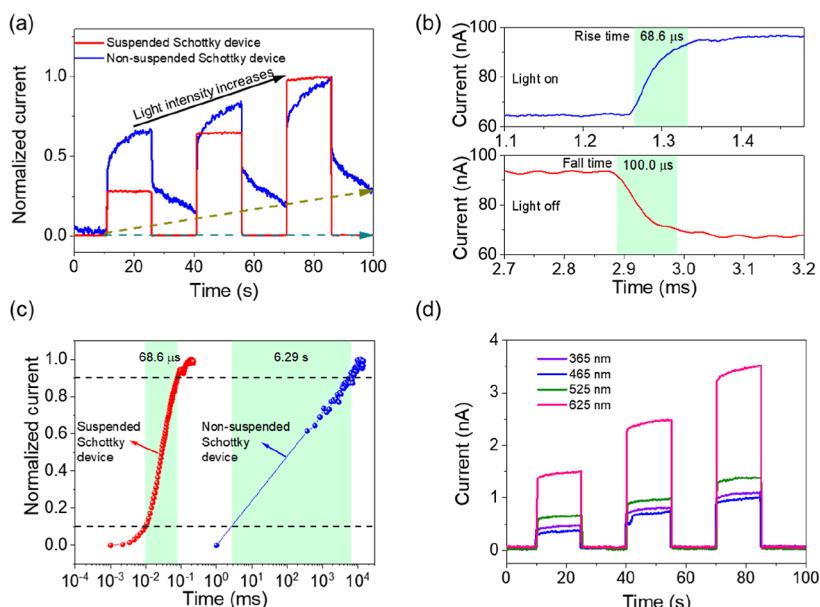
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**Figure 1.** (a) Optical images and schematic diagrams of nonsuspended (top) and suspended (bottom) MoS<sub>2</sub> devices. The scale bar is 20  $\mu\text{m}$ . (b) AFM image of a suspended MoS<sub>2</sub> device, with the line scan along the black and red dashed lines shown on the bottom. (c) Photoluminescence spectra and (d) Raman shifts of the suspended and nonsuspended MoS<sub>2</sub>.



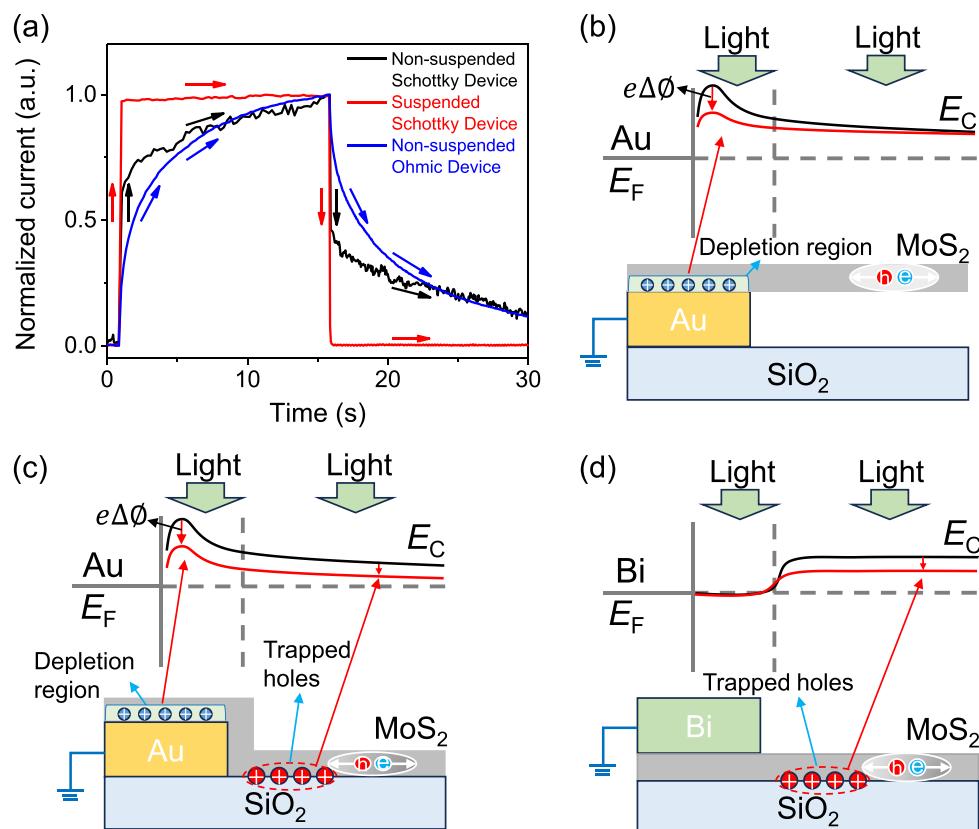
**Figure 2.** (a) Normalized current of the nonsuspended and suspended Schottky-contact devices under increased light illumination over time, as shown by the black arrow. (b) Transient measurements showing the current rise and fall processes of the suspended device when the light turns on or off. (c) Summary of the photocurrent rise time of the suspended and nonsuspended devices. (d) Current of the suspended device under different wavelengths of light illumination with increased intensity over time.

suspended device remains stable within a wide range of light intensity. Furthermore, according to the comparison of photoresponses of MoS<sub>2</sub>-based photodetectors with different structures, a detailed analysis of the photoresponse mechanisms has been performed. This work provides fundamental guidance for the design of 2D photodetectors with high performance.

## RESULTS AND DISCUSSION

The slow speed of light response and decay of responsivity with light intensity have become obstacles for the further development of MoS<sub>2</sub>-photodetectors, which is mainly related

to the dangling bonds or slow traps on the surface of the oxide underneath. Therefore, to avoid such influence on the MoS<sub>2</sub> channel and improve the light response speed, we design photodetectors with a fully suspended MoS<sub>2</sub> channel and compare them with the normal nonsuspended MoS<sub>2</sub> devices. The optical images and schematic diagrams of the non-suspended (on the top) and suspended (on the bottom) MoS<sub>2</sub> field effect transistors are shown in Figure 1a, with both MoS<sub>2</sub> flakes in contact with and on top of the Au electrodes. The nonsuspended devices are fabricated by lithographically patterning the Au/Cr electrodes on the p<sup>+</sup>-Si/300 nm SiO<sub>2</sub> wafer, followed by mechanical exfoliation and dry-transfer of

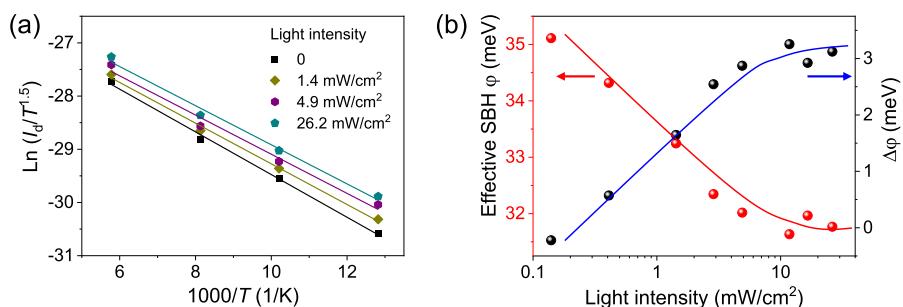


**Figure 3.** (a) Comparison of normalized current of a suspended Schottky-contact device, nonsuspended Schottky-contact device, and nonsuspended Ohmic-contact device. Schematic energy band structure and device structure of the (b) suspended Schottky-contact device, (c) nonsuspended Schottky-contact device, and (d) nonsuspended Ohmic-contact device under light illumination.

MoS<sub>2</sub>.<sup>23</sup> To achieve complete separation of the Si substrate and the MoS<sub>2</sub> channel, the fabrication procedures of the substrate with suspended MoS<sub>2</sub> are modified. Au/Cr metals are first evaporated on the p<sup>+</sup>-Si/300 nm SiO<sub>2</sub> wafer without photolithography, so that the metal film covers the full surface of the wafer. Then the surface microtrench is formed by photolithography and reactive ion etching of the Au/Cr, SiO<sub>2</sub>, and Si in the channel area, leaving a ~1  $\mu$ m deep microtrench. Then a MoS<sub>2</sub> flake is mechanically exfoliated and dry-transferred onto the microtrench, forming the suspended MoS<sub>2</sub> channel. For both suspended and nonsuspended MoS<sub>2</sub>, the channels are ~10  $\mu$ m long.

To make sure the MoS<sub>2</sub> channel is fully suspended, the atomic force microscopy (AFM) characterization is conducted and the image is shown in Figure 1b. The black line and red line show the depth of the MoS<sub>2</sub> and blank channel along with the horizontal x axis direction, which shows that the channel depth is about 1  $\mu$ m and the MoS<sub>2</sub> channel is suspended as a flat membrane over the microtrench and separated from the substrate. The MoS<sub>2</sub> quality of the two devices is examined with photoluminescence (PL) and Raman spectroscopy. The slight red-shift of the suspended MoS<sub>2</sub> compared with the nonsuspended MoS<sub>2</sub> indicates tensile strain after the suspension (Figure 1c). As shown in Figure 1d, the  $E_{2g}^1$  and  $A_{1g}$  Raman peaks also show red-shift for the suspended MoS<sub>2</sub> (378.4 and 404.7  $\text{cm}^{-1}$ ) compared with the nonsuspended device (382.2 and 406.6  $\text{cm}^{-1}$ ), further confirming the tensile strain in the suspended MoS<sub>2</sub>. The frequency differences of the two devices are 26.3 and 24.4  $\text{cm}^{-1}$ , which shows the two MoS<sub>2</sub> flakes are more than six layers.<sup>24</sup>

Then the electrical properties of the two devices are explored. Figure S1a shows the  $I_d$ – $V_{ds}$  curves of the suspended (blue) and nonsuspended (red) devices, both of which exhibit obvious nonlinearity. This behavior is quite common for 2D transistors with Schottky junctions between Au and 2D semiconductors, which is mainly caused by Fermi level pinning effect,<sup>25–27</sup> and strain between metal and 2D semiconductors.<sup>28,29</sup> Transfer characteristics of the two devices are plotted in Figure S1b. The suspended device shows smaller on/off ratio than the nonsuspended device, which might be related to the thicker dielectric layer or varied contact condition. Then the optoelectronic properties of the two devices over a long time scale are studied under the illumination of a commercial 525 nm LED. As shown in Figure 2a, both devices have a strong response to light. For the nonsuspended device, when the light is turned on, the current experiences an abrupt jump and then begins to slowly increase. The rise time, which is defined by the time needed to increase from 10% to 90% of the maximum current, is extracted as ~6 s from the light response curve for the nonsuspended device. By comparison, the current of the suspended device only experiences an abrupt increase after the light is turned on and then reaches saturation. Obviously, the suspended device shows a much faster response speed compared to the nonsuspended device, which agrees well with our expectation. Moreover, after the light is turned off, the current of the suspended device can rapidly drop back to its initial value while the current of the nonsuspended device remains at a much higher value than the initial value and gradually decreases. As the light intensity ( $P_{\text{light}}$ ) increases, the dark current of the nonsuspended device keeps increasing



**Figure 4.** (a) Arrhenius plots of the suspended MoS<sub>2</sub> device under light illumination ( $\lambda = 525$  nm) of different intensities. (b) Effective SBHs and SBH reduction as a function of light intensity.

while that of the suspended device can always drop back to the baseline of dark current.

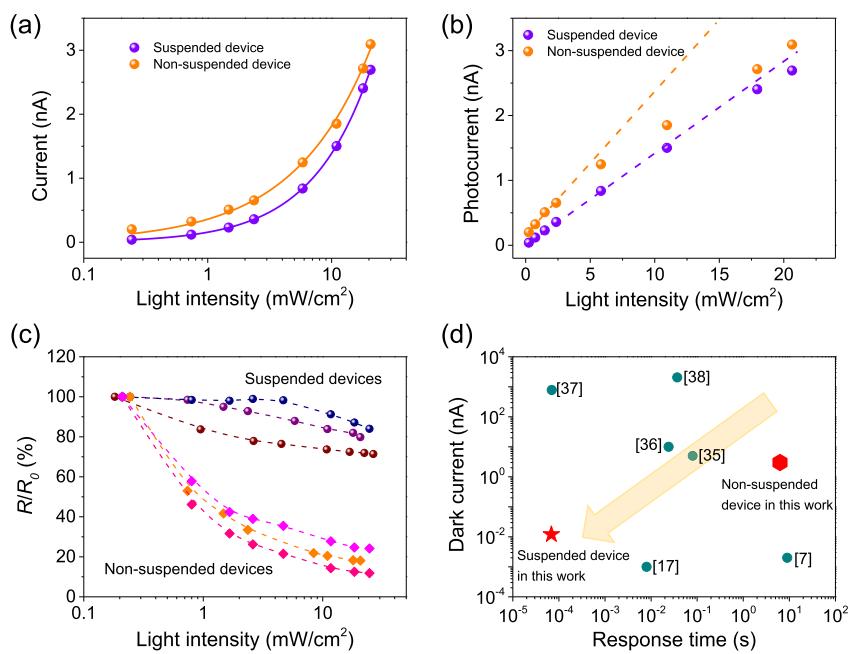
To obtain the specific light response time of the suspended device, transient photocurrent measurements within the short time scale are carried out with a 405 nm pulsed laser with a rise and fall time less than 3 ns, and the transient current rising and falling processes are shown in Figure 2b. The rise and fall time are extracted as 68.6 and 100.0  $\mu$ s. Then the rising process of the two devices is plotted in Figure 2c, which shows the rise time of suspended device is reduced by about 5 orders of magnitude compared to the nonsuspended device. To further determine the fast response of the suspended device, the photoresponse cutoff frequency has been measured. As shown in Figure S2, the cutoff frequency at  $-3$  dB is about 64 kHz, indicating an ultrafast response speed. Then the responses to light with long time scale at different wavelengths for the suspended device are measured using four LEDs (365, 465, 525, and 625 nm). As shown in Figure 2d, no matter which wavelength of LED is used, the suspended device exhibits ultrafast response speed. Moreover, the device shows the strongest response to the 625 nm light, which shows that the device has the highest quantum efficiency under 625 nm light illumination.

According to the results above, when the suspended device is exposed to light, its current will undergo a rapid increase in tens of microseconds and then reach saturation, where there is only a rapid current rising process, as shown in Figure 3a. Similarly, when the light is turned off, there is only a rapid current falling process. In contrast, for the nonsuspended device, when the light is turned on, its current will undergo a rapid increase and then a slow increase on the time scale of a few seconds. The situation when light is turned off is similar. From previous models, the photocurrent of the phototransistors can be mainly generated from the photoconductive effect, photogating effect, and photoinduced Schottky barrier reduction.<sup>30–32</sup> The photoconductive effect directly generates photocurrent in the device channel, the photogating effect mainly occurs at the interface between the MoS<sub>2</sub> channel and SiO<sub>2</sub> substrate, and the photoinduced Schottky barrier reduction occurs in the contact area of the MoS<sub>2</sub> and metal electrodes. Since the MoS<sub>2</sub> in the suspended device is fully separated from the SiO<sub>2</sub> substrate, the photogating effect is suppressed in this case. Therefore, the results suggest that the photocurrent generation in the channel (by the photogating effect and photoconductive effect) is a slow process due to trap states (on the time scale of seconds), while the photocurrent generation by photoinduced Schottky barrier reduction is a fast process (on the time scale of microseconds).

To further validate the hypothesis, a device with Ohmic contact is fabricated by using Bi as the contact layer. The rigidly linear  $I$ – $V$  curve (Figure S3) shows this device has a desirable Ohmic contact. When the light is turned on, the current begins to grow slowly, and it takes several tens of seconds to saturate (Figure 3a). Compared with the suspended device and the nonsuspended Schottky-contact device, the nonsuspended Ohmic-contact device only has a slow photocurrent rising process. When the light is turned off, the current also only undergoes a slow falling process. The photocurrent of the nonsuspended Ohmic-contact device mainly comes from the photogating effect in the channel because there is no Schottky barrier. Therefore, the results are consistent with the hypothesis above.

To better understand the photoresponse mechanisms of these three types of devices, the schematic energy band diagram and device structure of the suspended Schottky-contact device, nonsuspended Schottky-contact device, and nonsuspended Ohmic-contact device under light illumination are shown in Figure 3b–d, respectively. For the suspended device, due to the Schottky junction formed at the contact area between MoS<sub>2</sub> and Au, its current is governed by a dual-Schottky-junction model.<sup>33</sup> When the device is under light illumination, photogenerated electron–hole pairs will narrow the depletion region in the MoS<sub>2</sub> side and reduce the Schottky barrier height. As the Schottky barrier is reduced, device current can be improved according to the dual-Schottky junction model, which is the origin of the photocurrent and exhibits a fast response speed.

For the nonsuspended Schottky-contact device, its current is also governed by the dual-Schottky-junction model due to the Au–MoS<sub>2</sub> contact. Similarly, this device also undergoes a fast reduction of the Schottky barrier under light illumination. Besides, the photogating effect plays a role at the same time, and the photogenerated holes are trapped by the impurities on the oxide surface and distribute spatially in the channel, which forms an additional electric field just like the effect of gate voltage, and enhances the channel conductance. Due to the prolonged excess carrier lifetime and the gradual accumulation of hole trapping, the conductance modulation by the photogating process is a slow process. For the nonsuspended Ohmic-contact device, the conduction energy band of MoS<sub>2</sub> is below its Fermi level near the Bi electrode, resulting in a negligible Schottky barrier. Therefore, there is only a photogating effect under light illumination, which will slowly enhance the channel conductance. These mechanisms can well explain the dynamic photoresponse of different types of MoS<sub>2</sub>-based photodetectors.



**Figure 5.** (a) Total current of the suspended and nonsuspended MoS<sub>2</sub> devices under light illumination dependent on light intensity ( $P_{\text{light}}$ ) and fitting results with eq 2. (b) Photocurrent ( $I_{\text{ph}} = I_{\text{illum}} - I_{\text{dark}}$ ,  $I_{\text{illum}}$  is the device current under light illumination and  $I_{\text{dark}}$  is the dark current) of the suspended and nonsuspended MoS<sub>2</sub> devices dependent on light intensity. The dashed linear lines are guides to the eye. (c) The photoresponsivity changes of the device as light intensity increases. (d) A summary of the response time and dark current of the photodetectors based on a pure MoS<sub>2</sub> channel in literature.

To further validate the photoresponse mechanism of the fully suspended device, temperature-dependent photoresponse measurements are carried out and the light-intensity-dependent Arrhenius plots ( $\ln(I_{\text{ds}}/T^{1.5})$  vs.  $1000/T$ ) are shown in Figure 4a. The Arrhenius plots with negative slopes confirm that the device has a Schottky contact. As the light intensity increases, the slope gradually increases, showing a gradually decreasing Schottky barrier height. Then the Schottky barrier height and its reduction are extracted and plotted as a function of the light intensity in Figure 4b. As the light intensity increases from 0 to 26.2 mW/cm<sup>2</sup>, the Schottky barrier height gradually decreases from 34.9 to 31.8 meV. These results are well consistent with the hypothesis above.

Previously, a photoresponse model based on Schottky-contact MoS<sub>2</sub> phototransistors has been established.<sup>33</sup> According to this model, the relationship between  $P_{\text{light}}$  and Schottky barrier reduction  $\Delta\phi$  has been written as

$$P_{\text{light}} = P_{\text{light}}^S \left[ \exp\left(\frac{\Delta\phi}{mkT}\right) - 1 \right] \quad (1)$$

where  $P_{\text{light}}^S$  is defined as the critical light intensity given by  $P_{\text{light}}^S = \frac{\hbar\omega n_i}{2\alpha\tau_0}$ , in which  $\hbar\omega$  is the photon energy,  $n_i$  is the intrinsic electron concentration (per unit area) of MoS<sub>2</sub>,  $\alpha$  is the light absorption ratio by MoS<sub>2</sub>, and  $\tau_0$  is the minority carrier recombination lifetime in MoS<sub>2</sub>.  $M$  is the ideality factor determining how effectively the potential barrier is reduced by light illumination, which depends on the device structure, light absorption, defects in the junction, etc. The total current  $I_{\text{illum}}$  is related with the light intensity  $P_{\text{light}}$  as follows

$$I_{\text{illum}} = I_{\text{ph}} + I_{\text{dark}} = I_{\text{dark}} \left( \frac{P_{\text{light}}}{P_{\text{light}}^S} + 1 \right)^m \quad (2)$$

As shown in Figure 5a,  $I_{\text{illum}}$  of the suspended and nonsuspended devices are plotted as a function of  $P_{\text{light}}$ . Since the two devices are both predominant by the Schottky junction, their  $I_{\text{illum}}$  can be well fitted with eq 2. From the fitting results, the  $P_{\text{light}}^S$  of the suspended and nonsuspended devices are  $1.08 \times 10^{-5}$  and  $2.85 \times 10^{-5}$  mW/cm<sup>2</sup>, respectively. According to the definition of  $P_{\text{light}}^S = \frac{\hbar\omega n_i}{2\alpha\tau_0}$ , the minority carrier recombination lifetime of the suspended MoS<sub>2</sub> is a few times shorter than that of the nonsuspended MoS<sub>2</sub>. This result is consistent with the experimental data described above. The ideality factors are extracted as 0.90 and 0.70 for the suspended and nonsuspended devices, respectively. The near unity ideality factor of the suspended device shows it is more effective to reduce the Schottky barrier by light illumination than the nonsuspended device. To further prove this,  $I_{\text{ph}}$  of the suspended and nonsuspended devices are extracted and plotted as a function of  $P_{\text{light}}$  (Figure 5b). While the  $I_{\text{ph}}$  of the suspended device shows a near linear dependence on  $P_{\text{light}}$ , the  $I_{\text{ph}}$  of the nonsuspended device gradually saturates as the  $P_{\text{light}}$  increases from 0.24 to 20.6 mW/cm<sup>2</sup>.

It is notable that the suspended device shows better stability of responsivity  $R$  ( $R = I_{\text{ph}}/(A \times P_{\text{light}})$ ,  $A$  is the device area) with varying light intensity than the nonsuspended device. As shown in Figure 5c, the normalized responsivities  $R/R_0$  ( $R_0$  is the responsivity with an initial light intensity of  $\sim 0.18$  mW/cm<sup>2</sup>) of three suspended and three nonsuspended devices are plotted as a function of  $P_{\text{light}}$ . When the light intensity increases from 0.18 to 26.2 mW/cm<sup>2</sup>, the  $R$  values of nonsuspended devices decrease to about 20% of the initial value, while those of suspended devices only decrease to about 80%. For nonsuspended devices, the photocurrent originates from the combination of light-induced Schottky barrier reduction and photogating, which makes its initial  $R$  larger than that of the

suspended device (Figure S4), but photocurrent from photogating has an obvious decay with the increase of light intensity due to the gradually filled trap states in the channel, which is quite common for such devices. As shown in Figure S5, the linear dynamic range (LDR) of the suspended device is determined using two additional devices by measuring the photocurrents over a larger range of light intensities, which shows high linearity of photocurrent at different light intensities, and leads to the excellent LDR of 66 and 54 dB. We further measure the noise power spectral densities of these two devices and extract the detectivity of the suspended phototransistors (Figure S6).

In addition, low dark current is also crucial for low-noise photodetectors.<sup>34</sup> However, it is generally difficult to simultaneously achieve a high response speed and low dark current. In Figure 5d, we summarize the dark current and response time (rise time) of photodetectors based on a pure MoS<sub>2</sub> channel.<sup>7,17,35–38</sup> The closer the data point is to the origin (lower left corner) of the coordinate axis, the better the device performance is. For the nonsuspended device in this work, it has a few seconds of response time and a relatively large dark current, making it far from ideal. For the suspended device, due to the existence of Schottky barrier and the separation of the MoS<sub>2</sub> channel oxide, it has very fast response speed (68.6  $\mu$ s) and extremely low dark current (12 pA), which makes it closer to the origin than other reported devices and shows excellent performance.

While 2D heterojunction photodiodes also show high speed,<sup>39–42</sup> suspended 2D phototransistors support gate tuning of photodetection performance such as the photo-responsivity. As shown in Figure S7, by varying gate voltage, the responsivity can be tuned from  $1.4 \times 10^{-2}$  to 20.9 A/W, by more than 3 orders of magnitude at a light intensity of 2.6 mW/cm<sup>2</sup>. Furthermore, the responsivity reaches 138.4 A/W at 0.21 mW/cm<sup>2</sup>, which is much higher than that for 2D heterojunction photodiodes. The results show that the suspended phototransistor has the potential to achieve both high responsivity and high speed. Furthermore, the fully suspended structure allows us to reveal the intrinsic photo-responses of the 2D phototransistor without the substrate effect. The performance of the suspended 2D photodetectors can serve as a benchmark for interface-engineered 2D photodetectors.

## CONCLUSION

In summary, photodetectors with high response speed and stable responsivity are prepared based on fully suspended 2D MoS<sub>2</sub>. Due to the complete separation of the MoS<sub>2</sub> channel and Si substrate underneath, the photogating effect is effectively suppressed in this device. Then the photocurrent rise time and fall time are measured as 68.6 and 100.0  $\mu$ s by transient photocurrent measurement, respectively, which are about  $10^5$  times faster than the nonsuspended devices. Moreover, the suspended devices exhibit stable responsivity within a large range of light intensity, which shows great potential for practical applications in photodetectors. A detailed analysis of the photoresponse mechanisms of MoS<sub>2</sub>-based photodetectors is performed by designing devices with different structures and comparing their photoresponses. The results in this work can provide a fundamental understanding of photoresponse mechanisms of MoS<sub>2</sub>-based photodetectors and guide the design of high-performance 2D photodetectors.

## ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsphotonics.5c00146>.

Fabrication and measurement of suspended and non-suspended MoS<sub>2</sub> photodetectors, basic *I*–*V* characteristics without light illumination, cutoff frequency measurement, LDR measurement, noise measurement, gate tuning of responsivity, and measured responsivity at varying light intensity (PDF)

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### Author Contributions

<sup>†</sup>J.W. and Z.L. made an equal contribution.

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## Notes

The authors declare no competing financial interest.

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