Supplementary Materials for

High Performance Molybdenum Disulfide Amorphous Silicon Heterojunction Photodetector

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Device Fabrication and Measurement:

Flakes of $MoS₂$ were exfoliated on $SiO₂$ substrates from a piece of $MoS₂$ crystal (provided by the SPI Supplies, www.2spi.com) using the scotch-tape mechanical cleavage method. Subsequently, the thin film $MoS₂$ flakes were coated with amorphous silicon (a-Si) thin films, deposited by plasma-enhanced chemical vapour deposition (PECVD) using silane gas source at 260 C. Metal contacts were formed by the conventional lift-off technique, where Ti (20 nm) and Au (160 nm) were deposited by electron-beam evaporation at room temperature. Current-voltage characteristics of devices were measured by an Agilent 4155C semiconductor parameter analyzer, under dark and illuminated conditions. For the latter, a fiber-coupled broadband light out of a halogen lamp was used. The optical power density was about 10 mW/cm². We also measured photoresponse of devices at three wavelengths corresponding to blue, green, and red colors. For these measurements, we used standard LEDs as light sources and the incident power was about 0.4 mW/cm². The thickness of the $MoS₂$ flakes was measured using a mechanical, stylus-based step profiler, Alpha-Step IQ Surface Profiler. Figure S1 shows the top view

photomicrograph of the hybrid a-Si/MoS₂ metal-semiconductor-metal (MSM) photodetector and a lateral line scan of the MoS_2 flake, showing that the thickness of flake is about 60 nm.

Figure S1: **a**, top view photomicrograph of the a-Si/MoS₂ MSM photodetector. **b**, lateral line scan of the $MoS₂$ flake, showing that the thickness of flake is about 60 nm.

Measurement Setup for Transient Photoresponse:

Figure S2 shows the schematic of the setup used for measurements of the transient photoresponse of devices. The LED light source was biased by a 100 Hz square voltage out of a function generator. The light pulse was incident on the device under test (DUT) from top side. A diffuser was used to make sure the light intensity is uniform across a wide area of several cm^2 , thus making sure the DUT is receiving uniform light intensity. The distance between the LED and the device was about 4 cm to obtain uniform illumination. The DUT was biased by a DC voltage to extract photo current pulses. The photocurrent was amplified by a current to voltage (I to V) amplifier to obtain a voltage output. Finally, the voltage signal was fed to an oscilloscope along with the reference (sync) signal out of the function generator.

Figure S2: schematic of the setup used for measurements of the transient photoresponse of devices.

High Dark Current of metal-semiconductor-metal (MSM) photodetectors with just thin film MoS₂:

In the beginning of this research, we studied thin film $MoS₂$ as an active layer for MSM photodetectors. Figure S3a and b show the schematic of the device cross section and the top view photomicrograph of the fabricated device, respectively. The device length and width is about 5 and 6 µm, respectively. The current-voltage (IV) characteristics of the device were measured under dark and illuminated conditions. For the latter, a fiber-coupled broadband light out of a halogen lamp was used. The optical power density was about 10 mW/cm². Figure S3c shows the IV characteristics.

Figure S3: **a**, schematic of cross section of the MoS_2 MSM device, and **b**, the top view photomicrograph of the fabricated device. **c**, dark and photo current-voltage (IV) characteristics. For the latter, a fiber-coupled broadband light out of a halogen lamp was used. The optical power density was about 10 mW/cm².

As seen, the drawback of the MSM device, with just $MoS₂$ active layer, is the high dark current. For example, at 1V applied voltage, the dark current is about 40 pA. This value is about two orders larger than the dark current of the $a-Si/MoS₂$ device presented in the main text. The high dark current results in a low dynamic range which limits the application of $MoS₂ MSM$ photodetector in practical systems. Therefore, we found that a-Si layer helps to reduce the dark current, see the discussions in the main text. From Fig. S3c, one may also notice that both photo and dark IV curves are asymmetric with respect to the applied voltage, as the currents for identical positive and negative biases are not the same. For example, the photo currents are about 1nA and 40pA at +1 and -1V, respectively. This could be due to some variations or asymmetry in the Schottky barriers between the metal contacts and the $MoS₂$ layer.

Responsivity of Devices with 300 nm thick a-Si:

To test what happens for a thicker a-Si, we made another set of devices with 300 nm a-Si with the same dimensions as those of the first set , reported in the main text, (Note that the diffusion length of a-Si is about 300 nm). Supplementary Fig. S4 shows the photoresponsivity data measured under similar conditions. As seen, devices with 300 nm a-Si, with and without $MoS₂$, give the same photoresponse for all three incident lights. Therefore, in this case, photogenerated electrons completely reside inside the a-Si layer.

Figure S4: photoresponsivity of a-Si/MoS₂ and a-Si MSM devices with 300 nm thick a-Si.

Analysis of the transient response:

By using the data from Figs 3c and 3d, in the main text, we have obtained some statistical information on the transient responses of the a-Si and the a-Si/MoS₂ hybrid devices. The data are summarized in Figures S5-7. Figures S5 and S6 show the fall times for the two devices where we considered ten pulses from Figs 3c and 3d. To estimate the response time, we found the time it takes to switch from one level (e.g. high level) to reach about 90% of the other level (e.g. low level).

In addition, the transient response of the hybrid device can be modeled with an exponential function, $y = y_0 + A \exp(-t/t_0)$, where t_0 is the time constant. The statistical distribution of the time constant is shown in Fig. S7. It should be noted that due to the residual conductivity, the transient response of the a-Si device could not be modeled with an exponential function.

Figure S5: Statistical distribution of the fall time of the transient response of the $a-Si/MoS₂$ device.

Figure S6: Statistical distribution of the fall time of the transient response of the a-Si device.

Figure S7: Statistical distribution of the time constant (t_0) of the transient response of the a-Si/MoS₂ device. Extracted by fitting the pulses with the exponential decay function, $\exp(-t/t_0)$.

Using the transient response measurment setup, we measured the transient photoresponse of the two sets of devices, four different devices in total. Those are devices with 100 and 300 nm a-Si, with and without $MoS₂$. The applied DC voltage was 1V for most of measurements, except for devices with just 100 nm a-Si, where we had to increase the applied voltage to 5-10 V to obtain a less noisy signal and to make the output signal amplitude comparable to that of the hybrid a- $Si/MoS₂$ device. The results are summarized in the main text, Figs. 3c and 3d for devices with 100nm a-Si, and in Supplementary Figures S8-9 for devices with 300 nm a-Si. For the two devices with 300 nm amorphous silicon, with and without $MoS₂$, one can see that the transient responses are virtually identical, see Figs. S8 and S9. We estimated rise and fall times of about 2- 3 msec for these two devices, irrespective of the presence of the $MoS₂$ flake. Therefore, $MoS₂$ is not functioning in the hybrid device with 300 nm a-Si. In other words, once the amorphous silicon is thicker than the diffusion length of electrons in amorphous silicon, then electrons tend to move laterally inside the top a-Si layer without crossing the junction and being transferred to the $MoS₂$ layer.

Figure S8: Transient photoresponse of the $a-Si/MoS₂$ device with 300 nm $a-Si$ for three wavelengths corresponding to blue, green, and red wavelengths. The curves are shifted vertically for display purposes.

Figure S9: Transient photoresponse of the a-Si MSM device with 300 nm a-Si for three wavelengths corresponding to blue, green, and red wavelengths. The curves are shifted vertically for display purposes.

Photovoltaic effect in the fabricated devices:

From Fig. 2c, in the main text, one can see that there is some photovoltaic (PV) effect in the photo IV characteristics, especially the a-Si device shows a larger PV effect. This is due to process variations related to Schottky barriers. We also observed photovoltaic effect in $MoS₂$ samples. Supplementary Fig. S3 and Fig. S10 show PV effect in $MoS₂$ and a-Si/MoS₂ MSM devices, respectively.

Figure S10: Dark and Photo IV characteristics of two a- $Si/MoS₂$ MSM devices. The photo IV shows some photovoltaic (PV) effect, and it varies due to process variations related to Schottky barriers.

Denoising transient data of the a-Si device:

The transient response of the a-Si device, shown in Fig. 3d of the main text, contains some high frequency noise. We used data averaging to obtain cleaner, less noisy traces. Supplementary Figure S11 shows three denoised traces when we apply 10, 15, and 20 point averaging algorithm. Figure S11(a) shows several pulses and part (b) shows a zoomed-in transition. As seen, while data averaging helps to denoise the pulses, the timing for the persistent photoconductivity of a-Si is the same for all traces. As shown on Figure S11(b), a 3msec transient response is obtained for

the persistent photoconductivity. Thus, averaging only helps to minimize the high frequency noise and does not affect the slow transition due to the persistent conductivity.

Figure S11: (a) transient response of the a-Si device denoised by averaging. (b) a zoomed-in transition, showing that averaging only helps to minimize the high frequency noise and does not affect the slow transition (~3msec) due to the persistent conductivity.