

## Supporting Information

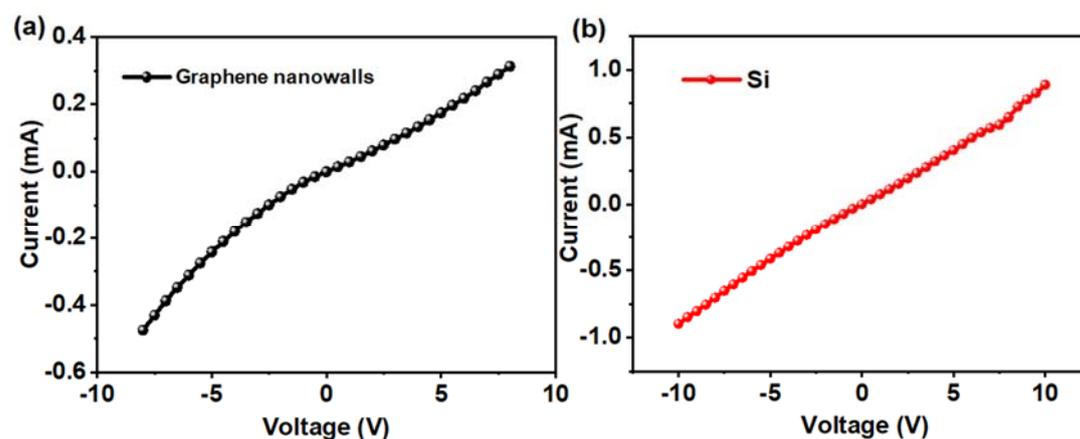
### High Performance Graphene Nanowalls/Si Self-powered Photodetectors with HfO<sub>2</sub> as an Interfacial Layer

Yuheng Shen<sup>1,2</sup>, Yulin Li<sup>1,2</sup>, Wencheng Chen<sup>1,2</sup>, Sijie Jiang<sup>1</sup>, Cheng Li<sup>1</sup> and Qijin Cheng<sup>1,2,\*</sup>

<sup>1</sup>) School of Electronic Science and Engineering, Xiamen University, Xiamen, 361102, P. R. China.

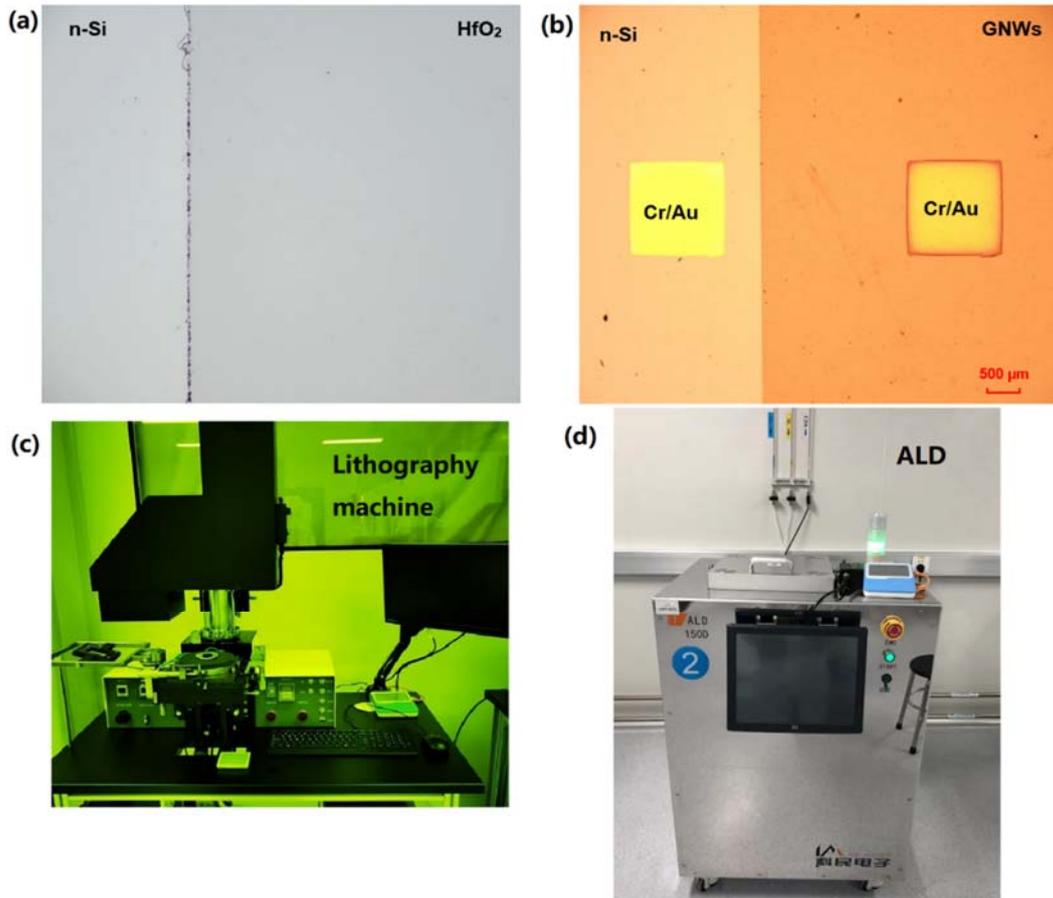
<sup>2</sup>) Shenzhen Research Institute of Xiamen University, Shenzhen, 518000, P. R. China.

\*Correspondence: E-mail: qijin.cheng@xmu.edu.cn

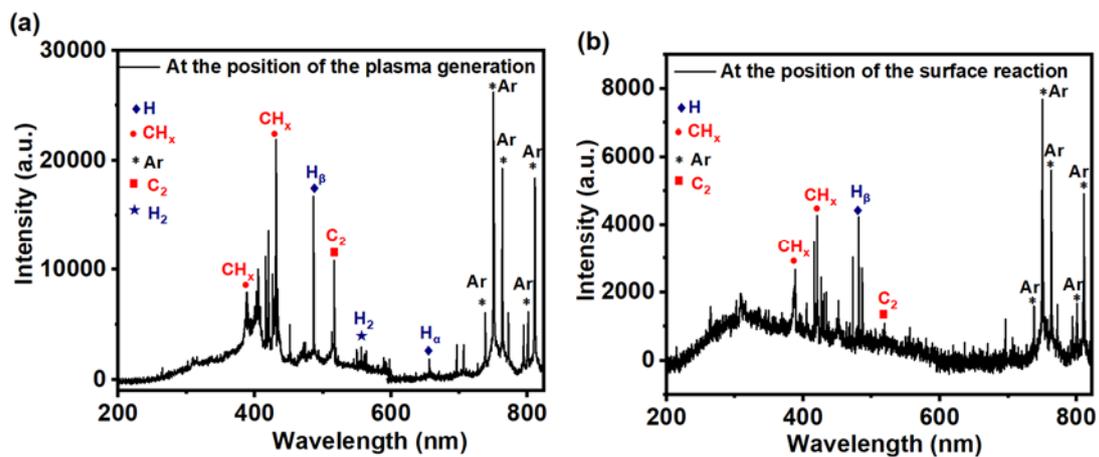


**Figure S1.** Ohmic contact of (a) GNWs and (b) Si.

The current-voltage characteristic curves for Cr/Au-GNWs-Cr/Au and Cr/Au-Si-Cr/Au are shown in Figure S1a and S1b, respectively. The almost linear I-V characteristic curves indicate that quasi ohmic contact is formed.



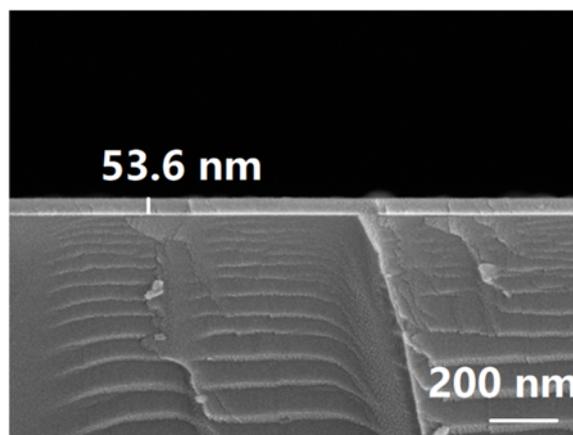
**Figure S2.** (a) Photograph of the Si substrate with a 3 nm HfO<sub>2</sub> layer (the two-thirds area of the Si substrate was covered with a 3 nm HfO<sub>2</sub> layer). (b) Photograph of the GNWs/HfO<sub>2</sub>/Si photodetector. (c) Photograph of lithography equipment. (d) Photograph of ALD equipment.



**Figure S3.** OES spectra at different positions of the remote PECVD system: (a) at the

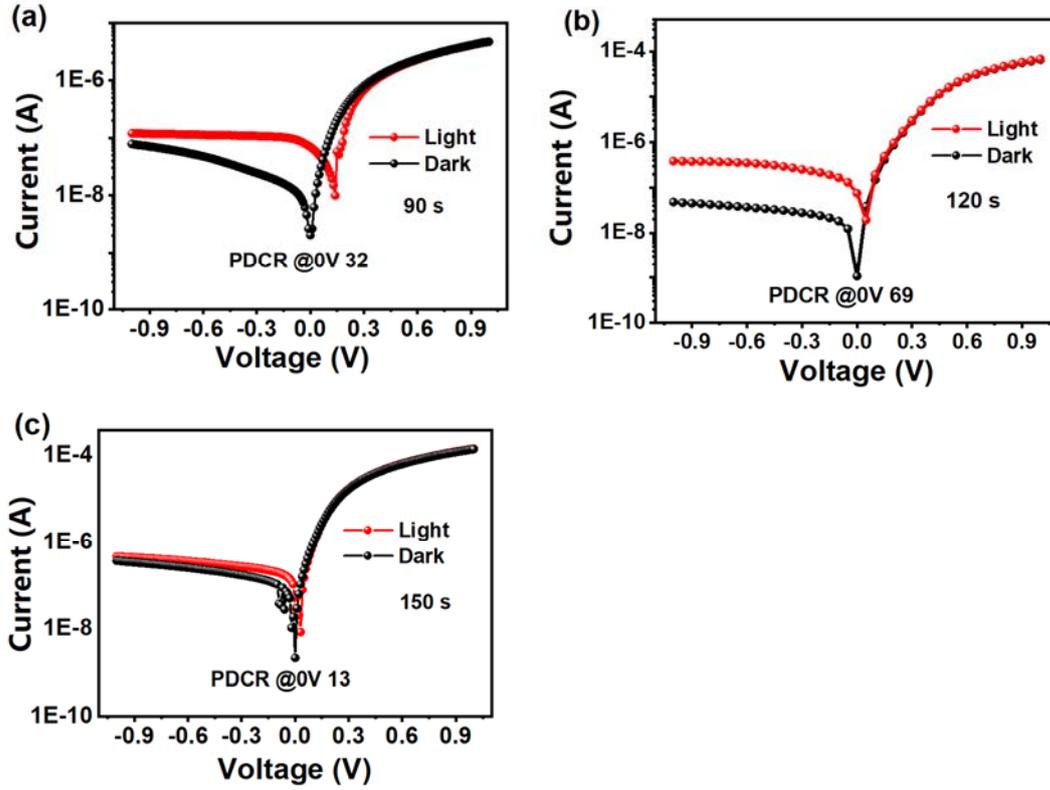
position of the plasma generation and **(b)** at the position of the surface reaction.

Figure S3 shows OES spectra at different positions of the remote PECVD system in the growth process of GNWs under the condition of a CH<sub>4</sub>/Ar flow rate ratio of 10 sccm/40 sccm, an RF power of 300 W, a growth temperature of 900 °C and a growth time of 120 s. As shown in Figure S3a, a series of spectral lines, including C<sub>2</sub>, CH<sub>x</sub>, Ar, H<sub>α</sub>, H<sub>β</sub>, H<sub>2</sub>, etc., can be observed at the position of the plasma generation. Notably, Figure S3a exhibits several argon spectral lines with a high intensity, located in the 750-820 nm range. In contrast, Figure S3b does not show the spectra lines of H<sub>α</sub> and H<sub>2</sub> at the position of the surface reaction, which is favorable for the decrease of the etching effect by H-related radicals. Moreover, the intensity of argon spectral lines decreases at the position of the surface reaction, which can effectively reduce the impact of the unwanted ion bombardment.



**Figure S4.** A cross-sectional SEM image of HfO<sub>2</sub> grown on the Si substrate for 500 cycles.

As shown in Figure S4, the thickness of HfO<sub>2</sub> is approximately 53.6 nm and the growth rate is calculated to be 1.07 Å/cycle.

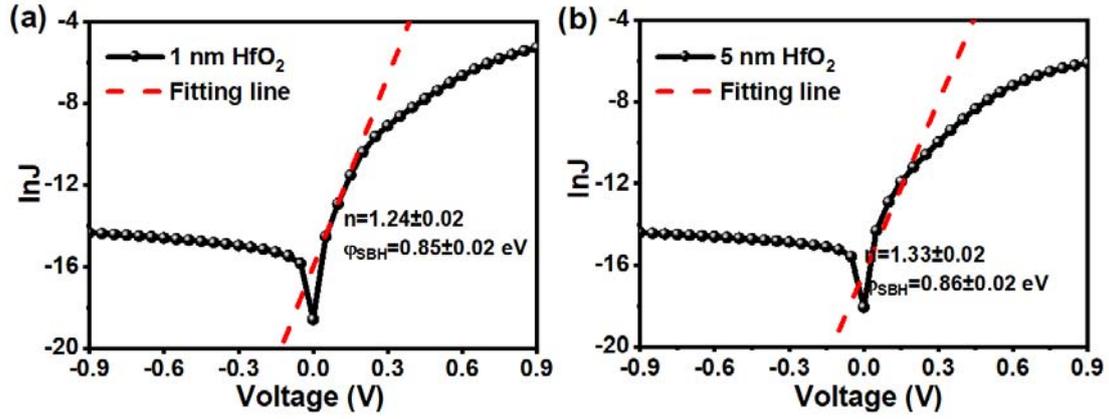


**Figure S5.** I-V characteristic curves of the GNWs/HfO<sub>2</sub>/Si photodetectors with different growth times of GNWs (a) 90 s, (b) 120 s, and (c) 150 s, respectively.

Figure S5 shows the I-V characteristic curves of the GNWs/HfO<sub>2</sub>/Si photodetectors with different growth times of GNWs. Table S1 lists the photoelectric parameters of the GNWs/HfO<sub>2</sub>/Si photodetectors with different growth times of GNWs. As shown in Table S1, the photodetector for GNWs grown for 120 s shows the best performance among all the fabricated devices.

**Table S1.** Photoelectric parameters of the GNWs/HfO<sub>2</sub>/Si photodetectors with different growth times of GNWs.

Time (s)	Current under dark condition (A)	Current under light condition (A)	PDCR @ 0V	Responsivity (A/W)
90	$2.02 \times 10^{-9} A$	$6.91 \times 10^{-8} A$	32	0.054
120	$1.07 \times 10^{-9} A$	$7.38 \times 10^{-8} A$	69	0.058
150	$2.16 \times 10^{-9} A$	$2.98 \times 10^{-8} A$	13	0.023



**Figure S6.**  $\ln J$ - $V$  curves of the GNWs/Si photodetectors with the thickness of (a) 1 nm and (b) 5 nm  $\text{HfO}_2$  layer.

Figure S6 shows the  $\ln J$ - $V$  curves of the GNWs/Si photodetectors with different thicknesses of  $\text{HfO}_2$  interface layer. We can obtain  $n$  and  $\varphi_{SBH}$  by fitting the linear part of the curve based on the equations (5) and (6). The results of the barrier height for photodetectors with 1 and 5 nm  $\text{HfO}_2$  are  $0.85 \pm 0.02$  and  $0.86 \pm 0.02$  eV, respectively. The results of the ideality factor for photodetectors with 1 and 5 nm  $\text{HfO}_2$  are 1.24 and 1.33, respectively.