

Supporting Information

High Performance Graphene Nanowalls/Si Self-powered Photodetectors with HfO₂ as an Interfacial Layer

Yuheng Shen^{1,2}, Yulin Li^{1,2}, Wencheng Chen^{1,2}, Sijie Jiang¹, Cheng Li¹ and Qijin Cheng^{1,2,*}

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

²⁾ Shenzhen Research Institute of Xiamen University, Shenzhen, 518000, P. R. China.

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

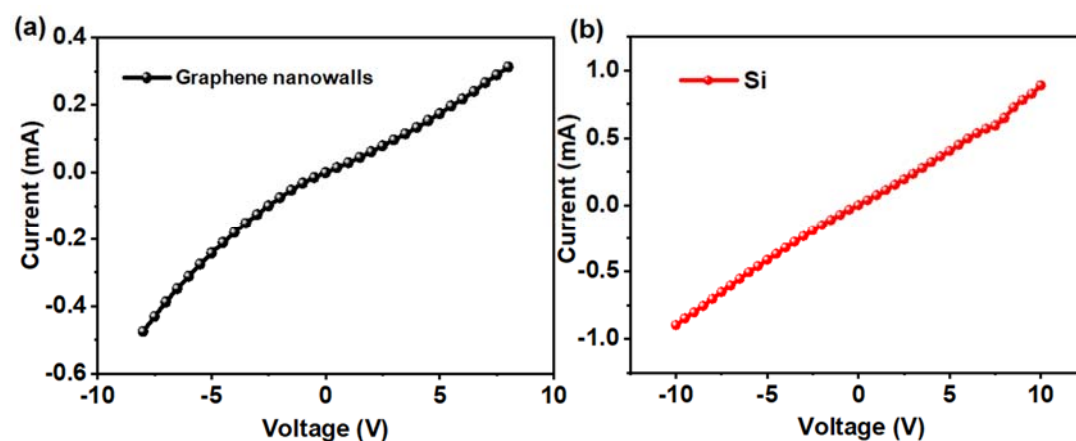


Figure S1. Ohmic contact of **(a)** GNWs and **(b)** Si.

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.

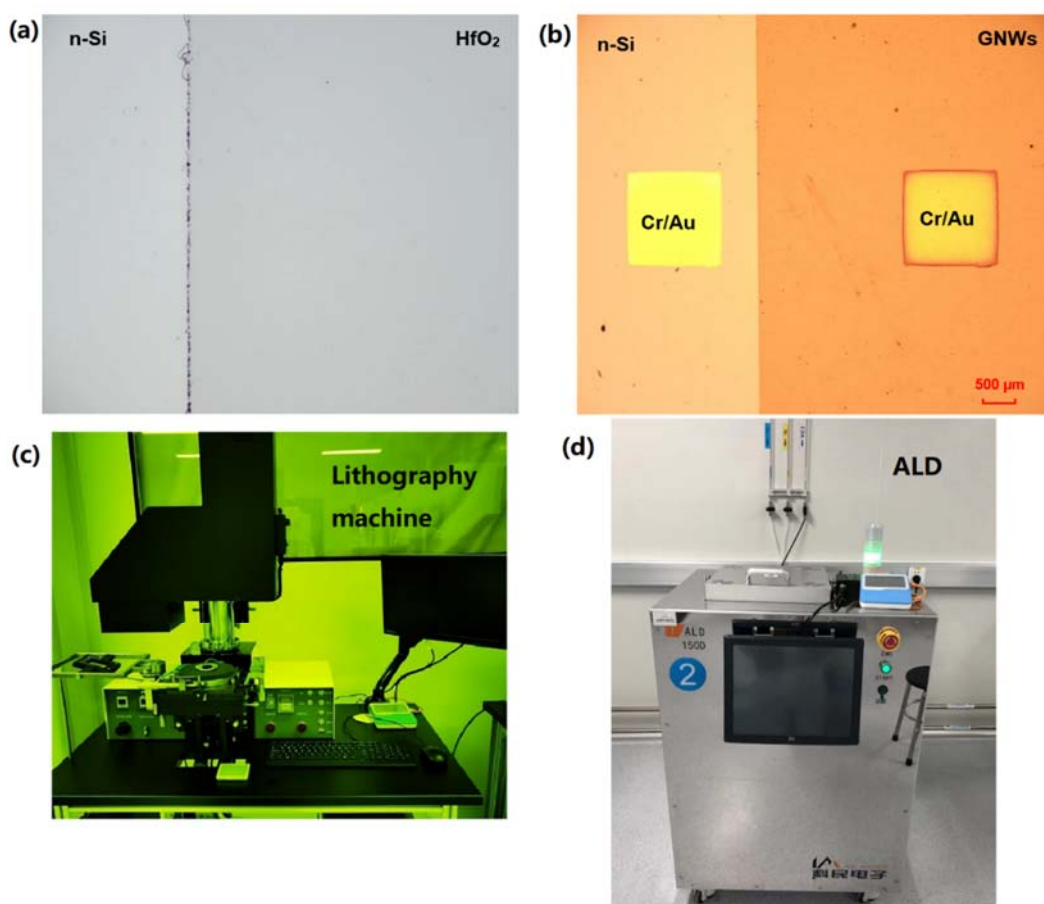


Figure S2. (a) Photograph of the Si substrate with a 3 nm HfO₂ layer (the two-thirds area of the Si substrate was covered with a 3 nm HfO₂ layer). (b) Photograph of the GNWs/HfO₂/Si photodetector. (c) Photograph of lithography equipment. (d) Photograph of ALD equipment.

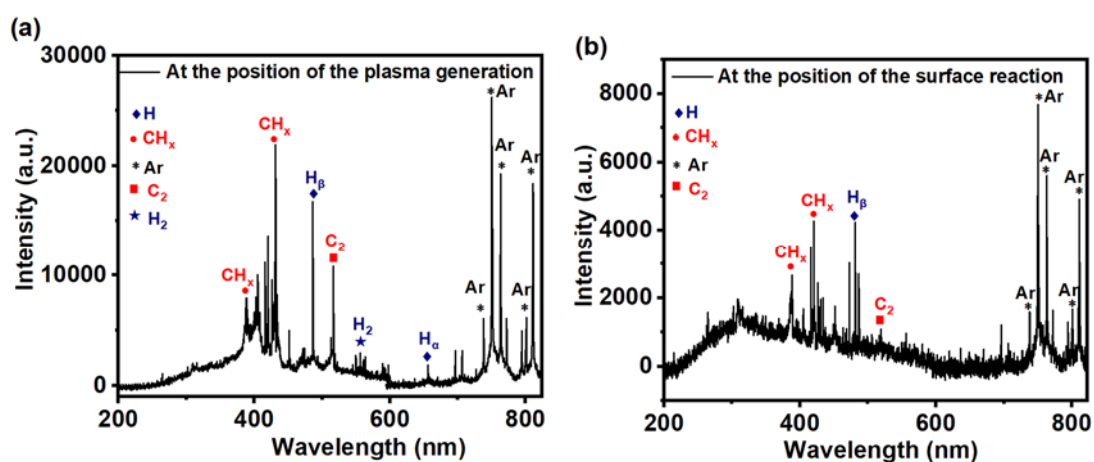


Figure S3. OES spectra at different positions of the remote PECVD system: (a) at the

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 CH₄/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 C₂, CH_x, Ar, H_α, H_β, H₂, 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_α and H₂ 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.

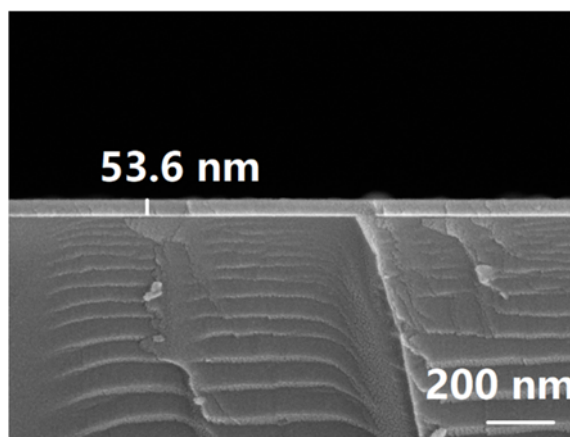


Figure S4. A cross-sectional SEM image of HfO₂ grown on the Si substrate for 500 cycles.

As shown in Figure S4, the thickness of HfO₂ is approximately 53.6 nm and the growth rate is calculated to be 1.07 Å/cycle.

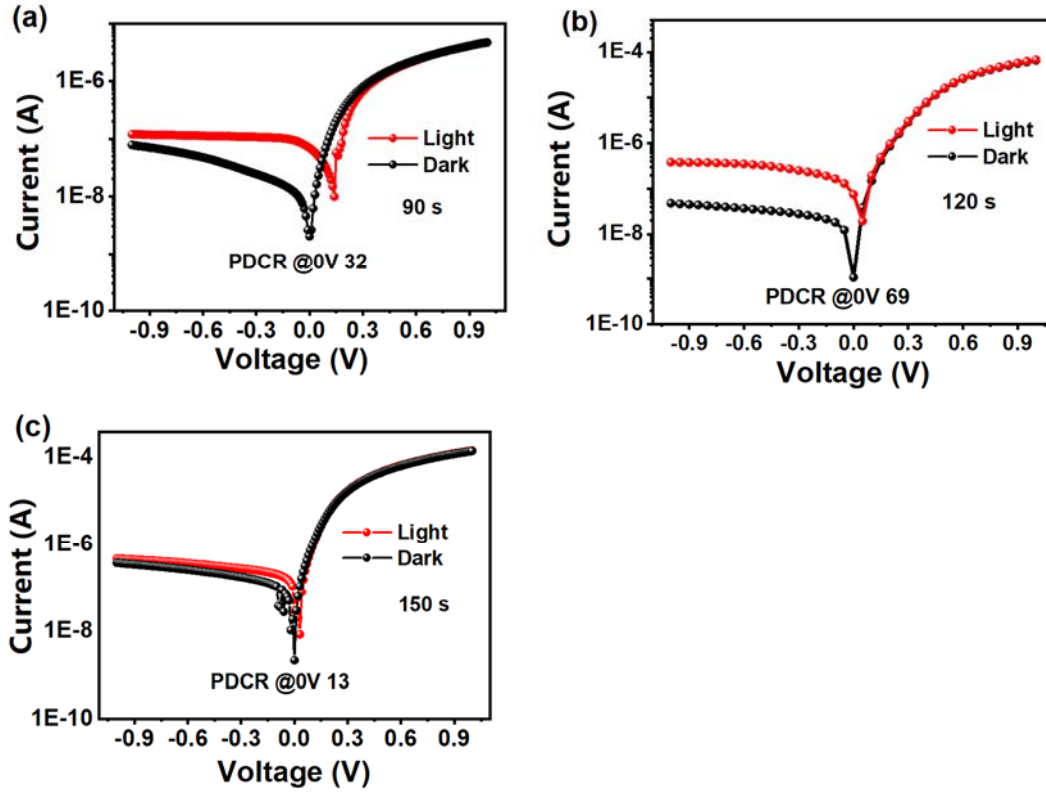


Figure S5. I-V characteristic curves of the GNWs/HfO₂/Si photodetectors with different growth times of GNWs (a) 90 s, (b) 120 s, and (c) 150 s, respectively.

Figure S5 shows the I-V characteristic curves of the GNWs/HfO₂/Si photodetectors with different growth times of GNWs. Table S1 lists the photoelectric parameters of the GNWs/HfO₂/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/HfO₂/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	$2.02 \times 10^{-9} \text{ A}$	$6.91 \times 10^{-8} \text{ A}$	32	0.054
120	$1.07 \times 10^{-9} \text{ A}$	$7.38 \times 10^{-8} \text{ A}$	69	0.058
150	$2.16 \times 10^{-9} \text{ A}$	$2.98 \times 10^{-8} \text{ A}$	13	0.023

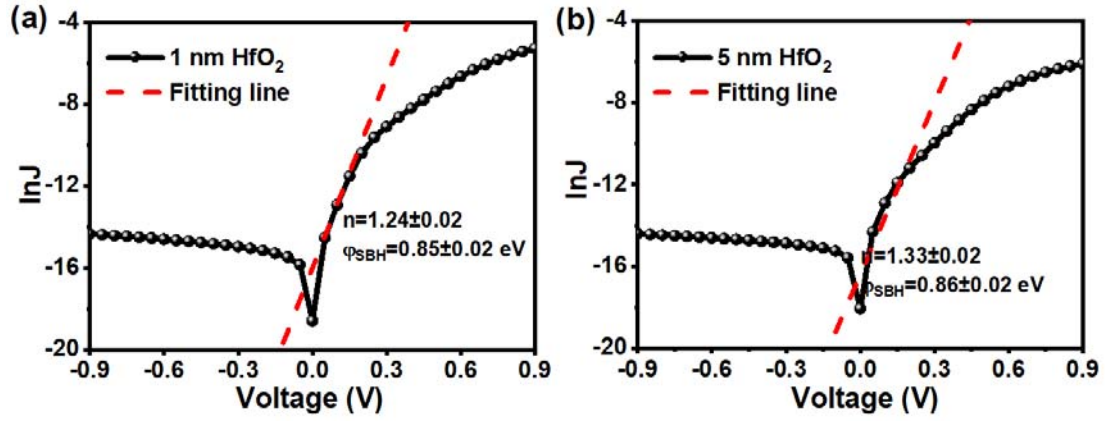


Figure S6. $\ln J$ -V curves of the GNWs/Si photodetectors with the thickness of (a) 1 nm and (b) 5 nm HfO₂ layer.

Figure S6 shows the $\ln J$ -V curves of the GNWs/Si photodetectors with different thicknesses of HfO₂ interface layer. We can obtain n and ϕ_{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 HfO₂ are 0.85 ± 0.02 and 0.86 ± 0.02 eV, respectively. The results of the ideality factor for photodetectors with 1 and 5 nm HfO₂ are 1.24 and 1.33, respectively.