Supplementary Information

Transition of Photoresponsivity in Graphene-Insulator-Silicon Photodetectors

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S1. Fabrication Process of MC-GIS Photodiodes and Experimental Set-up for Photoresponse Studies

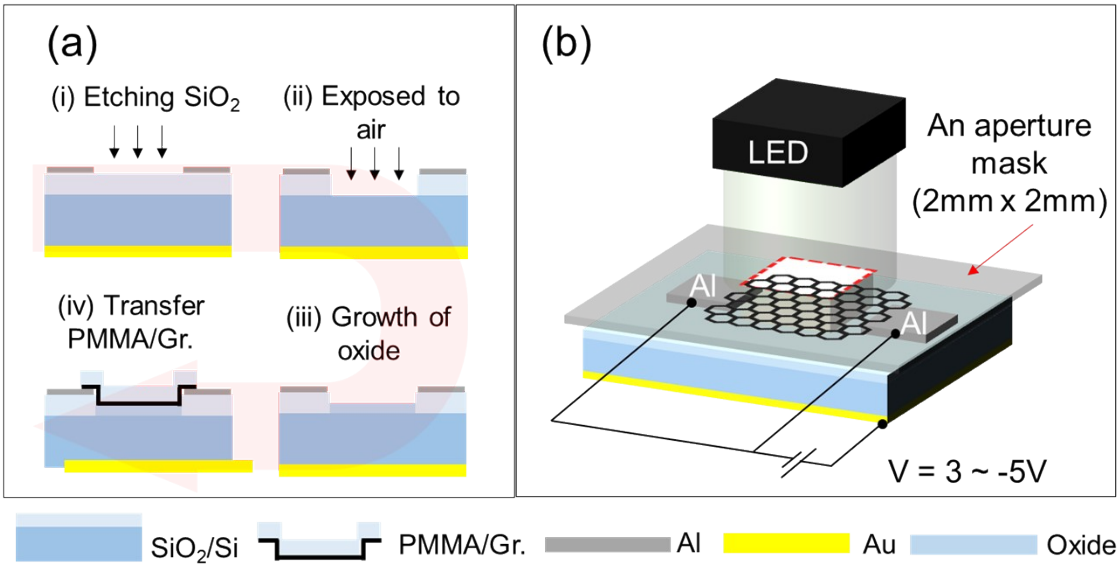

Figure S1. (a) Fabrication process of graphene-insulator-silicon (GIS) photodetectors. (b) Experimental setup for the electrical and optical response investigations of GIS photodetectors.

Figure S1a schematically shows the fabrication procedures of GIS photodiodes. The employed silicon wafer is p-type. **Figure S1b** shows experimental set-up for investing the photoresponse behaviors of GIS photodiodes.

S2. Optical Power-Dependent Capacitance-Voltage Characteristics

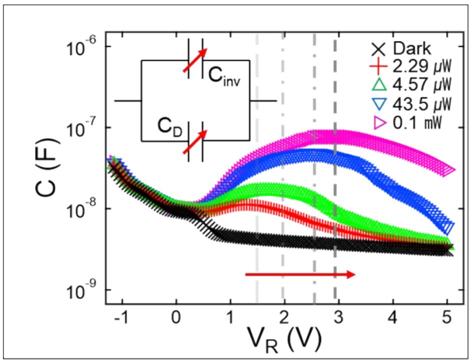

Figure S2. Capacitance-Voltage (C-V) characteristic curves as a function of the optical power (red light, 640 nm) measured at the modulation frequency of 1 kHz. The inset shows the schematics for parallel capacitance connection between the depletion region (C_D) and the inversion layer (C_{inv}).

Figure S2 illustrates the Capacitance-Voltage (C-V) characteristic curves obtained under dark conditions and as a function of red light optical power at a modulation frequency of 1 kHz.[1] The dark C-V curve indicates the absence of inversion at the reverse bias voltage, while a flat band is formed around a reverse bias voltage of 0.5 V.

The observed inversion behaviors in the C-V curves under high optical power illumination suggest the accumulation of photoelectrons at the SiO_2/Si interface. This is due to the higher photocarrier generation rate compared to the tunneling rate and recombination rate. The inset (top left) of Figure S3 presents the equivalent capacitance circuit for the semiconductor capacitance (C_s), which consists of the inversion layer capacitance (C_{inv}) and the depletion region capacitance (C_D) in parallel connection When a thin inversion layer is formed, the total semiconductor capacitance is primarily influenced by the inversion layer capacitance. With increasing optical power, the strongest (peak) inversion occurs at higher reverse bias voltages, attributable to the higher effective photoconductivity enhancement in silicon and the delayed expansion of the depletion width with higher optical power.

The non-linear behavior observed in the photocapacitance and photocurrent as a function of optical power and reverse bias voltage can be attributed to several factors, including the tunneling and recombination of charge carriers. With increasing optical power, the rate of photocarrier generation rises, resulting in a higher accumulation of photoelectrons at the SiO_2/Si interface and a stronger inversion. However, at higher optical powers, the tunneling rate and recombination rate of charge carriers also increase. This leads to a sub-linear increase in the inversion strength and a sub-linear increase in the photocurrent. Similarly, at higher reverse bias voltages, the tunneling rate becomes higher, which leads to a decrease in the inversion strength and a quasi-saturation of the photocurrent. Therefore, the behavior of the system exhibits non-linearity and is influenced by multiple factors such as optical power, wavelength of the light, and applied bias voltages.

S3. Photo Current-Voltage (I-V) Characteristics and Schottky Barrier Height Modulation

The current under light illumination (I_P) can be described using the modified Schottky emission theory, similar to the dark current shown in main manuscript, with the following equations.[2-4,5]

$$I_P = I_S^P \left\langle exp\left\{\left[q\left(V - I_P R_s\right)\right]/\eta^e k_B T\right\} - 1\right\rangle \quad \text{(S1)}$$

$$I_S^P = AA^* T^2 \exp\left(-q\Phi_s^P/k_B T\right) \exp\left(-\sqrt{\xi} d\right) \quad \text{(S2)}$$

$$\Phi_s^P(V) = \phi_0^P + q\Delta V_{ox}^P + q\Delta V_{Gr.}^P \quad \text{(S3)}$$

The net photocurrent (I_{ph}) can be obtianed by subtracting the dark current (I_D) from the current under light illumination (I_P) as below.

$$I_{ph} = I_P - I_D = \left(I_S^P\right)\left\langle exp\left\{\left[q\left(V - I_P R_s\right)\right]/\eta^e k_B T\right\} - 1\right\rangle - (I_S^D)\left\langle exp\left\{\left[q\left(V - I_D R_s\right)\right]/\eta^e k_B T\right\} - 1\right\rangle \quad \text{(S4)}$$

The effect Schottky barrier hight (Φ_s^P) under light illumination is expressed as

$$\Phi_s^P(V) = \Phi_s^D(V) + \Delta\Phi_s^P(V) \quad \text{(S5)}$$

Where $\Delta\Phi_s^P(V)$ is the SBH modulation induced by the light illumination.

Then

$$I_S^P = I_S^D \exp\left(-q\Delta\Phi_s^P/k_B T\right) \quad \text{(S6)}$$

$$I_P = I_S^D\left(\exp\left(-q\Delta\Phi_s^P/k_B T\right)\right)\left\langle exp\left\{\left[q\left(V - I_P R_s\right)\right]/\left(\eta^e k_B T\right)\right\} - 1\right\rangle \quad \text{(S7)}$$

Finally the net photocurrent is given by

$$I_{ph} = I_S^D\left(\exp\left(-q\Delta\Phi_s^P/k_B T\right)\right)\left\langle exp\left\{\left[q\left(V - I_P R_s\right)\right]/\left(\eta^e k_B T\right)\right\} - 1\right\rangle - I_S^D\left\langle exp\left\{\left[q\left(V - I_D R_s\right)\right]/\left(\eta^e k_B T\right)\right\} - 1\right\rangle \quad \text{(S8)]}$$

Further, equation S7 can be rearranged as below

$$\exp\left(q\Delta\Phi_s^P/k_B T\right) = \frac{I_S^D}{I_P}\left(exp\left\{\left[q\left(V - I_P R_s\right)\right]/\left(\eta^e k_B T\right)\right\} - 1\right) \quad \text{(S9-1)}$$

$$q\Delta\Phi_s^P/k_B T = ln\left(\frac{I_S^D}{I_P}\right) + ln\left(exp\left\{\left[q\left(V - I_P R_s\right)\right]/\left(\eta^e k_B T\right)\right\} - 1\right) \quad \text{(S9-2)}$$

Then the photoinduced SBH modulation ($\Delta\Phi_s^P$) can be obtained as below

$$\Delta\Phi_s^P = \frac{k_B T}{q}\left[ln\left(\frac{I_S^D}{I_P}\right) + ln\left(exp\left\{\left[q\left(V - I_P R_s\right)\right]/\left(\eta^e k_B T\right)\right\} - 1\right)\right] \quad \text{(S10)}$$

From the equation for dark currnet (I_D), I_S^D is expressed as below

$$I_S^D = I_D/\left(\left\langle exp\left\{\left[q\left(V - I_D R_s\right)\right]/\eta^e k_B T\right\} - 1\right\rangle\right) \quad \text{(S11)}$$

By inserting equation 11 into equation 10, the SBH modulation is reexpressed

$$\Delta\Phi_s^P = \frac{k_B T}{q}\left[ln\left(\frac{I_D/\left(\left\langle exp\left\{\left[q\left(V - I_D R_s\right)\right]/\eta^e k_B T\right\} - 1\right\rangle\right)}{I_P}\right) + ln\left(exp\left\{\left[q\left(V - I_P R_s\right)\right]/\left(\eta^e k_B T\right)\right\} - 1\right)\right] \quad \text{(S12-1)}$$

Further,

$$\Delta\Phi_s^P = \frac{k_BT}{q}\left[ln\left(\frac{I_D}{I_P}\right) - ln\left(\frac{\left(exp\left\{\left[q\left(V - I_PR_s\right)\right]/\left(\eta^e k_BT\right)\right\} - 1\right)}{\left(exp\left\{\left[q\left(V - I_DR_s\right)\right]/\eta^e k_BT\right\} - 1\right)}\right)\right] \quad \text{(S12-2)}$$

Here the ideality factor (η^e) is obtained by fitting the dark current (I_D) using Equation 1 in main manuscript. Finally we obtained the SBH modulation ($\Delta\Phi_s^P$) as a function of the experimental values of I_D and I_P as above.

Let's take a look at two cases as below.

Let $x = \left[q\left(V - I_PR_s\right)\right]/\left(\eta^e k_BT\right)$ or $x = \left[q\left(V - I_DR_s\right)\right]/\left(\eta^e k_BT\right)$

For x $\leq$ 1

$$e^x = 1 + x + \frac{x^2}{2} + \ \ldots$$

Then,

$$\Delta\Phi_s^P \approx \frac{k_BT}{q}\left[ln\left(\frac{I_D}{I_P}\right) - ln\left(\frac{\left[q\left(V - I_PR_s\right)\right]/\left(\eta^e k_BT\right)}{\left[q\left(V - I_DR_s\right)\right]/\left(\eta^e k_BT\right)}\right)\right]$$

$$= \frac{k_BT}{q}\left[ln\left(\frac{I_D}{I_P}\right) - ln\left(\frac{\left(V - I_PR_s\right)}{V - I_DR_s}\right)\right] \quad \text{(S13-1)}$$

This does not depend on the fitting parameter such as the ideality factor (η^e).

For x $\geq$ 1

$$\Delta\Phi_s^P \approx \frac{k_BT}{q}\left[ln\left(\frac{I_D}{I_P}\right) - ln\left(\frac{\left(exp\left\{\left[q\left(V - I_PR_s\right)\right]/\left(\eta^e k_BT\right)\right\}\right)}{\left(exp\left\{\left[q\left(V - I_DR_s\right)\right]/\eta^e k_BT\right\}\right)}\right)\right]$$

$$\Delta\Phi_s^P \approx \frac{k_BT}{q}\left[ln\left(\frac{I_D}{I_P}\right) - \frac{V - I_PR_s}{V - I_DR_s}\right] \quad \text{(S13-2)}$$

This also does not depend on the fitting parameter such as the ideality factor (η^e). With conjunction with dark current-voltage, the SBH modulation ($\Delta\Phi_s^P$) is extracted fully from the experimental current-voltage curves (I_P & I_D) as shown in main manuscript (Figure 5).

Finally the contribution of the second terms to the modulated SBH is neglible. Thus, the modulated SBH can be expressed as

$$\Delta\Phi_s^P \approx \frac{k_B T}{q}\left[ln\left(\frac{I_D}{I_P}\right)\right] \quad \text{(S14)}$$

References

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[5] S. M. Sze, K. K. Ng, *Physics of Semiconductor Devices, 3rd ed.*, John Wiley & Sons, Inc.: Hoboken, NJ, USA **2007**.