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## **Redefining Responsivity in Graphene-based Schottky Diodes**

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Graphene has great potential in flexible and rigid optoelectronic devices due to its atomic thickness (and hence flexibility), high conductivity and broadband transparency [1], [2]. Graphene forms a Schottky junction with semiconductor materials, which can be exploited as simple photodiodes [3], [4]. Recently, it was reported that insulated regions in graphene/silicon (G/Si) photodiodes, e.g. regions separated by oxides to allow contacting the graphene, have a significant contribution towards the extracted photocurrent [5], [6]. Based on these findings, we have optimized the design of graphene-based photodiodes. Here, we report on these graphene/silicon Schottky diodes with very high responsivity, including analysis of the spectral "fingerprint". Scanning photocurrent measurements (SPCM) support our claims as they provide local spatial distributions of photocurrents in the devices. Based on our experiments, we revisit the extraction of responsivity in previous literature, where the insulating regions have been largely ignored when calculating it. This leads to a distinct overestimation of responsivity.

n-type Si <100> wafers with a phosphorous doping concentration of  $2 \times 10^{14}$  cm<sup>-3</sup> and a thermally grown SiO<sub>2</sub> layer of 20 nm were used as substrates. The regions where graphene was to contact silicon were defined by photolithography and oxide was removed with buffered oxide etchant (BOE). Metal electrodes were patterned in a second photolithography step, followed by sputtering of 20 nm chromium (Cr) and 80 nm nickel (Ni) followed by the lift-off process. Chemical vapor deposited graphene was then transferred from copper foil onto the pre-patterned substrates with poly methyl methacrylate (PMMA) support. BOE was used to remove the native oxide on Si just before the graphene transfer. The PMMA was removed by baking at 180°C for 35 min, followed by acetone, isopropanol and drying. Finally, graphene regions were defined using photolithography and oxygen plasma etching. Devices with interdigitated SiO<sub>2</sub> and Schottky regions (PD1) and conventional Schottky diodes (PD2) were fabricated (Fig. 1).

Fig. 2 shows optical micrographs of the devices with dashed rectangles indicating regions covered by graphene. PD1 has larger oxide regions compared to PD2. Raman spectra of graphene on SiO<sub>2</sub>/Si and on Si indicate no significant differences in the quality of graphene (Fig. 3). Fig. 4 shows J-V characteristics of the diodes in the dark (solid lines) and under illumination (dashed lines) with a white LED light with a power density of 30  $\mu$ W/cm<sup>2</sup>. Both diodes are photosensitive and rectify with a ratio of  $3.9 \times 10^4$  and  $2.9 \times 10^4$  for PD1 and PD2, respectively. SPC measurements shown in Fig. 5 reveal that the regions of graphene on SiO<sub>2</sub> contribute significantly to the observed photocurrent in both diodes, confirming our previous results [4]. The absolute spectral response (SR) was measured by lock-in technique. In literature, the active diode area is assumed to be the same as the Schottky area ( $A_{G/Si}$ ). Historically this is reasonable, given that thick metal contacts have typically been used. However, the high transparency of graphene results in a significant contribution to the photocurrent from the Si under the G/SiO<sub>2</sub> region. This warrants redefining the active area to "generation area"  $A_g = A_{G/SiO_2} + A_{G/Si}$  (Fig. 6). Fig. 7 shows the SR of both diodes when considering only the Schottky area and the generation area. Very high responsivity of 0.54 A/W is observed for PD1 compared to 0.32 A/W for PD2 at a wavelength of 850 nm. The difference is attributed to more efficient collection of charge carriers in PD1. Ignoring the SiO<sub>2</sub> area can lead to significant overestimation of responsivity values, especially when the oxide area makes up a larger part of the device [7].

In summary, we demonstrated the significant contribution of insulating  $SiO_2$  regions towards photocurrents in G/Si based photodiodes. Considering this, we redefine the realistic photocurrent generation area, which should be used to calculate the actual responsivity obtained from certain graphene-based photodiodes.

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**Fig. 1:** (a) Schematic of G/Si photodiodes PD1 and PD2. In PD1, oxide region makes a significant part of the device compared to PD2.



**Fig. 2:** (a) Schematic of G/Si photodiodes PD1 and PD2. The dashed lines indicate the region covered by graphene.



Fig. 3: Raman spectra of graphene on Si (lower black) and on SiO<sub>2</sub>/Si region acquired using a 532 nm wavelength. Monolayer nature of CVD graphene is reflected by 2D/G intensity ratio > 1.



**Fig. 4:** Current density-voltage (J-V) plot of the diodes in the dark (solid lines) and under illumination (dashed lines) (white LED light, power density of  $30 \mu$ W/cm<sup>2</sup>.)



**Fig. 5:** Large-area scanning photocurrent maps of PD1 and PD2 showing the spatial distribution of photocurrent in the respective devices. Yellow regions, corresponding to oxide area, provide highest photocurrents.



**Fig. 6:** Schematic depicting the redefined active area as generation area in the present study compared to that used in conventional Schottky diodes using thick metal contacts.



**Fig. 7:** Responsivity vs. wavelength (lower x-axis) and energy (upper x-axis) of G/Si photodiodes for wavelengths ranging from 360 nm (3.4 eV) to 1200 nm (1 eV) at reverse biases of -2 V. The dashed lines represent responsivity calculated by considering the G/Si Schottky area only, while the solid lines are calculated realistically, i.e. considering the entire generation area. The black

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