Supporting Information

Graphene/Si Heterostructure with an Organic Interfacial LayerforSelf-Powered Photodetector with High ON/OFF Ratio

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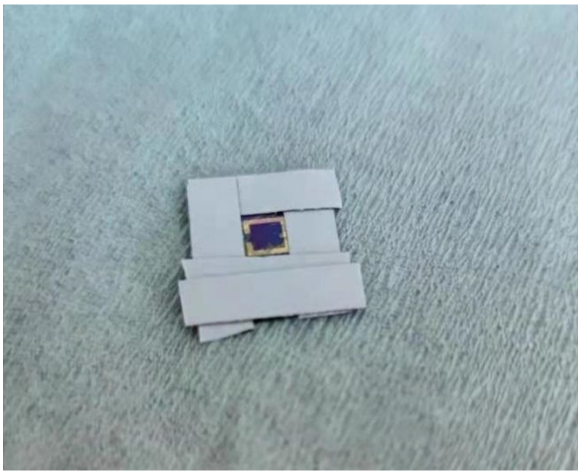

Figure S1. Optical image of fabricated Gr/Spiro-OMeTAD/Si device.

During the test, the area outside the electrode was covered to accurately define the device area.

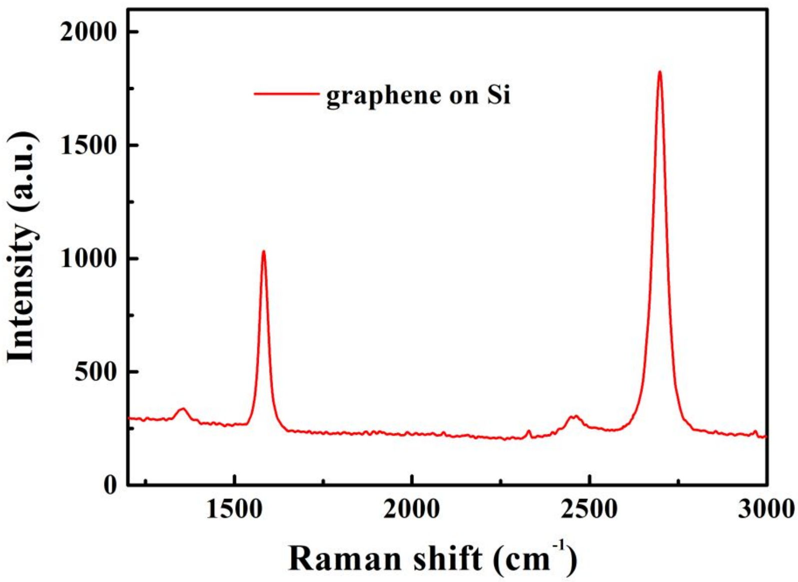

Figure S2. Raman spectrum of transferred graphene on Si substrate.

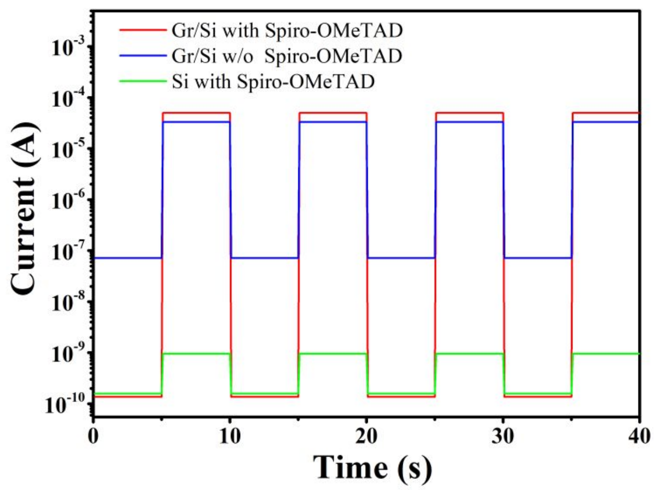

Figure S3.Measured photocurrent curves of Gr/Si device, Gr/Spiro-OMeTAD/Si device, and Spiro-OMeTAD/Si device at 532 nm illumination with 145 μW power with log scale ordinate.

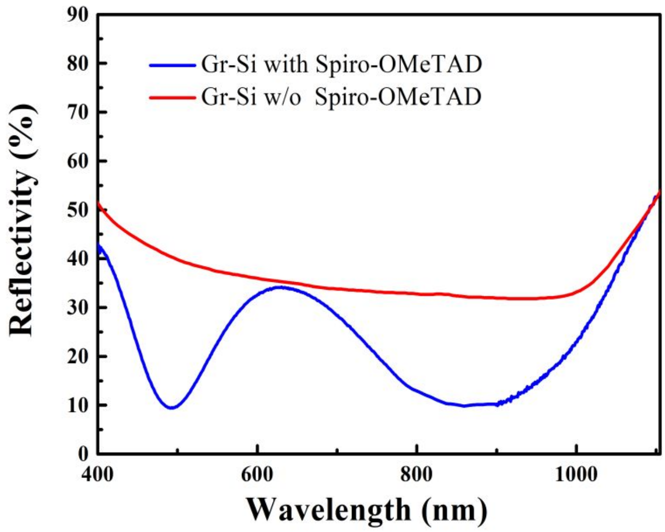

Figure S4. The light reflectance spectra of Gr/Si and Gr/Spiro-OMeTAD/Si PDs.

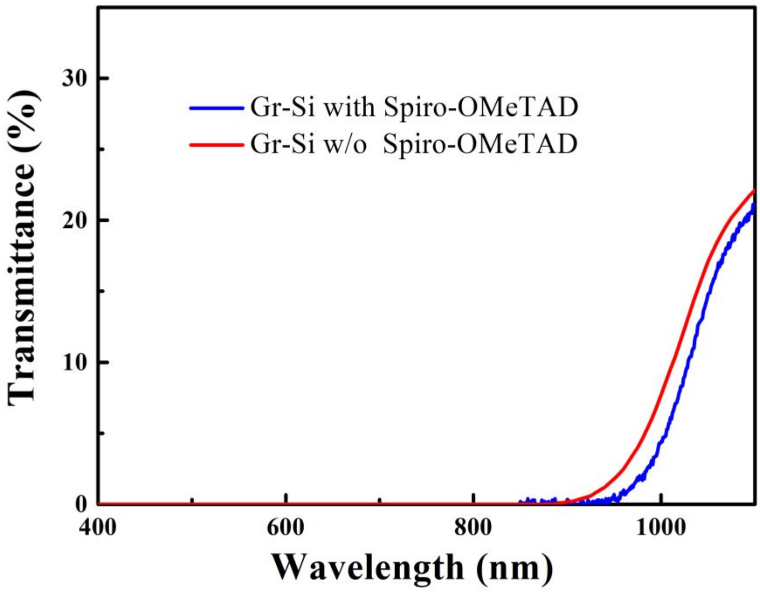

Figure S5. The light transmittance spectra of Gr/Si and Gr/Spiro-OMeTAD/Si PDs.

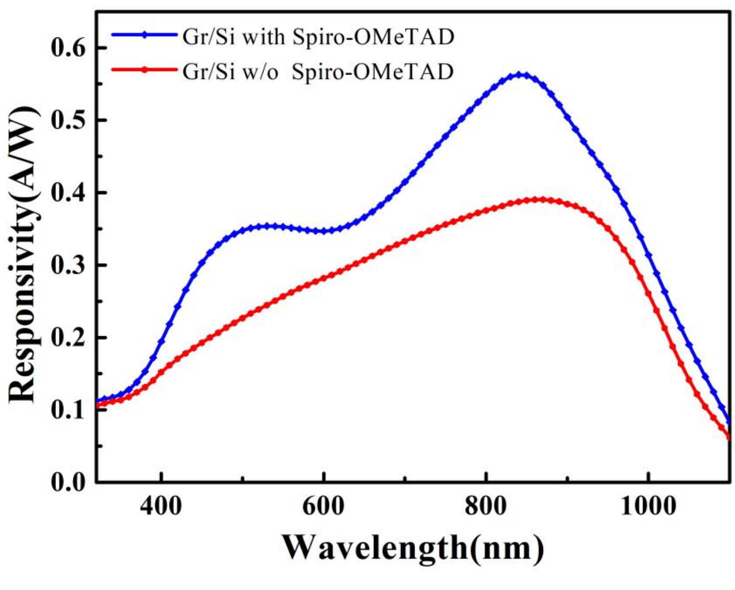

Figure S6. Responsivity spectrum with respect to wavelength of Gr/Si and Gr/Spiro-OMeTAD/Si PDs.

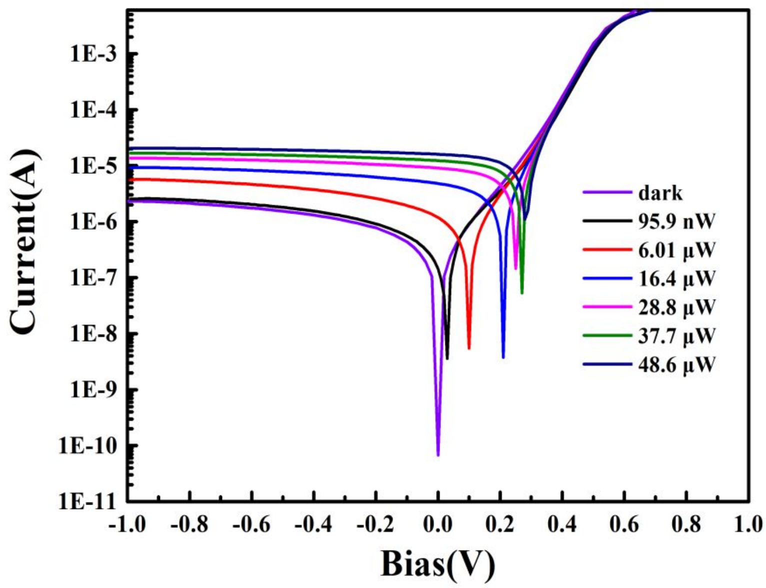

Figure S7. I-V curves recorded under light illumination with different power density graphene/Spiro-OMeTAD/Si PD.

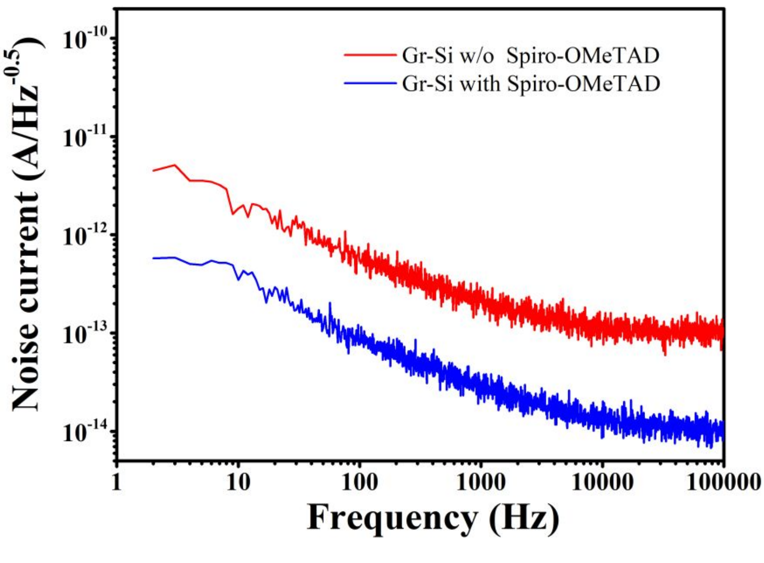

Figure S8.Frequency-dependent noise currentof Gr/Si PD and Gr/Spiro-OMeTAD/Si PD.

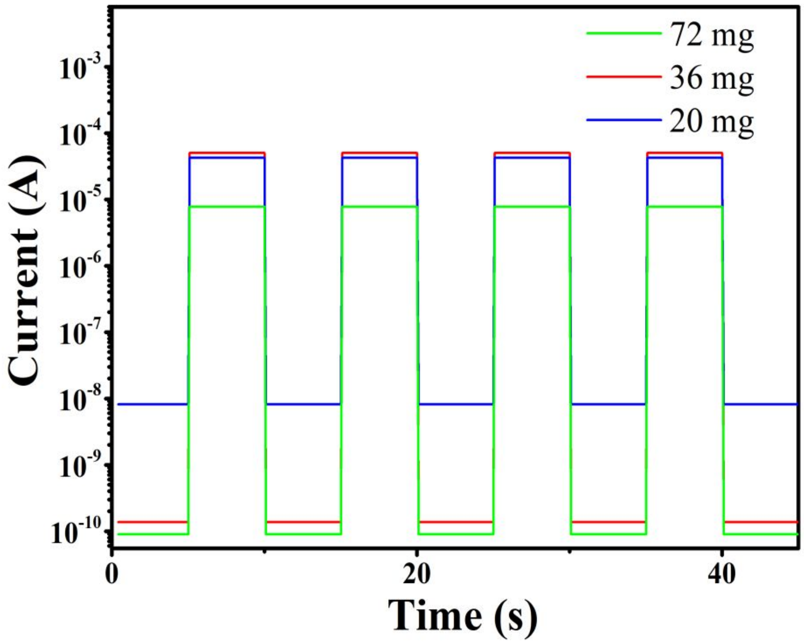

Figure S9.Measured photocurrent curves of the graphene/Spiro-OMeTAD/Si based device fabricated with different Spiro-OMeTAD solution concentration (72/36/20 mg/mL).

Table S1. A comparison of photodetector parameters

Ref.	Sample-type	Wavelength nm	Responsivity A/W	Speed τr/τf μs	Specific detectivity cm $Hz^{1/2}$/W	ON/OFF ratio
our	Gr/spiro-OMeTAD/Si	532	0.355	5.1/4.3	2.7×10^{11}	10^7
[1]	Gr/Si	850	0.435	1200/3000	7.69×10^9	10^4
[2]	Gr/Si	1550	0.0395	5/8	10^{11}	/
[3]	Gr/h-BN/Si	725	0.11	910/1080	2.83×10^{10}	10^7
[4]	Gr/WS_2/Si	690	8.96×10^4 (5V)	840/2100	8.86×10^{11}	/
[5]	Gr/GO/Si	633	0.65 (2V)	1000	1.88×10^{12}	2.73×10^5
[6]	GQDs/WSe_2/Si	740	0.707 (3V)	<200	4.51×10^9	10^4

7	$Gr/SiO_2/Si$	633	0.45	0.02/0.1	/	10^5
8	P3HT-graphene/Si	850	0.78 (1V)	4.7×10^6	2.6×10^{10}	/
9	Al2O3/Gr/Si	365	0.2	0.005	1.6×10^{13}	/
10	In_2S_3-nanoflake/Gr/Si	405	4.53×10^4 (2V)	33/40	3.02×10^{11}	666.1
11	Carbon-QDs/Gr/Si	600	0.29 (3V)	0.93/2.2	/	/
12	Si-QDs/Gr/Si	405	0.495	<0.025	7.4×10^9	/

(1) An, X.; Liu, F.; Jung, Y. J.; Kar, S. Tunable graphene-silicon heterojunctions for ultrasensitive photodetection. *Nano Lett* **2013,** *13* (3), 909-16, DOI: 10.1021/nl303682j.

(2) Wang, C.; Dong, Y.; Lu, Z.; Chen, S.; Xu, K.; Ma, Y.; Xu, G.; Zhao, X.; Yu, Y. High responsivity and high-speed 1.55 μm infrared photodetector from self-powered graphene/Si heterojunction. *Sensors and Actuators A: Physical* **2019,** *291*, 87-92, DOI: 10.1016/j.sna.2019.03.054.

(3) Won, U. Y.; Lee, B. H.; Kim, Y. R.; Kang, W. T.; Lee, I.; Kim, J. E.; Lee, Y. H.; Yu, W. J. Efficient photovoltaic effect in graphene/h-BN/silicon heterostructure self-powered photodetector. *Nano Research* **2020,** *14* (6), 1967-1972, DOI: 10.1007/s12274-020-2866-x.

(4) He, T.; Lan, C.; Zhou, S.; Li, Y.; Yin, Y.; Li, C.; Liu, Y. Enhanced responsivity of a graphene/Si-based heterostructure broadband photodetector by introducing a WS2 interfacial layer. *Journal of Materials Chemistry C* **2021,** *9* (11), 3846-3853, DOI: 10.1039/d0tc05796g.

(5) Wang, Y.; Yang, S.; Lambada, D. R.; Shafique, S. A graphene-silicon Schottky photodetector with graphene oxide interlayer. *Sensors and Actuators A: Physical* **2020,** *314*, DOI: 10.1016/j.sna.2020.112232.

(6) Sun, M.; Fang, Q.; Xie, D.; Sun, Y.; Qian, L.; Xu, J.; Xiao, P.; Teng, C.; Li, W.; Ren, T.; Zhang, Y.

Heterostructured graphene quantum dot/WSe2/Si photodetector with suppressed dark current and improved detectivity. *Nano Research* **2018,** *11* (6), 3233-3243, DOI: 10.1007/s12274-017-1855-1.

(7) Srisonphan, S. Hybrid Graphene–Si-Based Nanoscale Vacuum Field Effect Phototransistors. *ACS Photonics* **2016,** *3* (10), 1799-1808, DOI: 10.1021/acsphotonics.6b00610.

(8) Aydin, H.; Kalkan, S. B.; Varlikli, C.; Celebi, C. P3HT-graphene bilayer electrode for Schottky junction photodetectors. *Nanotechnology* **2018,** *29* (14), 145502, DOI: 10.1088/1361-6528/aaaaf5.

(9) Wan, X.; Xu, Y.; Guo, H.; Shehzad, K.; Ali, A.; Liu, Y.; Yang, J.; Dai, D.; Lin, C.-T.; Liu, L.; Cheng, H.-C.; Wang, F.; Wang, X.; Lu, H.; Hu, W.; Pi, X.; Dan, Y.; Luo, J.; Hasan, T.; Duan, X.; Li, X.; Xu, J.; Yang, D.; Ren, T.; Yu, B. A self-powered high-performance graphene/silicon ultraviolet photodetector with ultra-shallow junction: breaking the limit of silicon? *npj 2D Materials and Applications* **2017,** *1* (1), DOI: 10.1038/s41699-017-0008-4.

(10) Lu, J.; Zheng, Z.; Yao, J.; Gao, W.; Zhao, Y.; Xiao, Y.; Li, J. 2D In2 S3 Nanoflake Coupled with Graphene toward High-Sensitivity and Fast-Response Bulk-Silicon Schottky Photodetector. *Small* **2019,** *15* (47), e1904912, DOI: 10.1002/smll.201904912.

(11) Chen, X.; Yang, C.; Sun, H.; Ning, S.; Zhou, H.; Zhang, H.; Wang, S.; Feng, G.; Zhou, S. Enhanced photoresponsivity in carbon quantum dots-coupled graphene/silicon Schottky-junction photodetector. *Laser Physics Letters* **2019,** *16* (7), DOI: 10.1088/1612-202X/ab2040.

(12) Yu, T.; Wang, F.; Xu, Y.; Ma, L.; Pi, X.; Yang, D. Graphene Coupled with Silicon Quantum Dots for High-Performance Bulk-Silicon-Based Schottky-Junction Photodetectors. *Adv Mater* **2016,** *28* (24), 4912-9, DOI: 10.1002/adma.201506140.