A Broadband 3D Microtubular Photodetector Based on Single Wall Carbon Nanotubes-Graphene Heterojunction

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1. **The detailed fabrication process of the 3D microtubular photodetector based on single wall carbon nanotubes (SWCNTs) and graphene heterojunction**

The detailed fabrication process of the 3D microtubular photodetector based on SWCNTs/graphene heterojunction is shown below (Fig. S1):

(a) The silicon (Si) wafer was cleaned by dipping it in sulfuric acid (H_2SO_4) and hydrogen peroxide (H_2O_2) solution (volume ratio 4:1) at 85 °C for 20 minutes, followed by rinsing it with deionized water and drying it with nitrogen (N_2) gas. Then a layer of aluminum (Al) with the thickness of 50 nm was sputtered onto the top of Si wafer.

(b) The strained SiN_x layers were deposited on the Al sacrificial layer by a STS mixed frequency nitride plasma-enhanced chemical vapor deposition (PECVD) system, which consisted of a compressive trained SiN_x layer at the bottom and a tensile strained SiN_x layer on the top, with thicknesses of 80 nm and 50 nm, respectively. The compressive strained SiN_x layer was deposited at the conditions of 60 W, 380 kHz, 550 mTorr, 240 °C, 40.0 sccm of SiH_4 and 20 sccm of NH_3 (SiH_4/NH_3 ratio of 2). While the tensile SiN_x layer was deposited at the conditions of 30 W, 13.56 MHz, 900 mTorr, 240 °C, 40.0 sccm of SiH_4 and 55.0 sccm of NH_3 (SiH_4/NH_3 ratio of 0.72). N_2 flow was kept constant at 1960 sccm.. After the deposition process, reactive ion etching (RIE) process was carried out to pattern the SiN_x layer.

(c) A gate electrode consisting of chromium (Cr) and gold (Au) with the thicknesses of 10 nm and 50 nm, respectively, was deposited on the SiN_x layer, using magnetron sputtering and lift-off processes.

(d) A 30-nm-thick silicon dioxide (SiO_2) layer was deposited on the Cr/Au gate electrode to achieve buried gate structure, using PECVD process. Then, RIE etching process was performed to pattern the SiO_2 dielectric layer.

(e) A CVD-grown single-layer graphene (Trivial Transfer Graphene™, XFNANO Co. Ltd.) was transferred onto the top of the SiO_2 layer. Then, oxygen (O_2) plasma etching process was conducted to pattern the graphene layer.

(f) Source and drain electrodes consisting of 10-nm-thick Cr and 50-nm-thick Au were deposited and patterned on the top of the SiO_2 layer, using electron beam evaporation and lift-off processes. After this step, the planar (2D) graphene field-effect transistor (GFET) with buried gate structure was obtained.

(g) The SWCNTs ((6, 5) chirality enriched ($\geq$ 95%) semiconducting SWCNTs, Sigma-Aldrich) were dispersed in N-Methyl pyrrolidone (Aladdin) with a concentration of 0.1 mg/ml, followed by ultrasonication under the power of 800 W for 1 h. Then the resultant suspensions were centrifuged with 10,000 *g* for 1 h and the supernatant were collected. After that, drop the SWCNTs suspensions onto the surface of 2D GFET.

(h) After being drying out in an oven under 90 °C for 30 minutes, the 2D graphene/SWCNTs heterojunction photodetector was obtained.

(i) Etch the Al sacrificial layer selectively using HCl : H_2O (volume ratio is 2 : 1) solution and the highly strained SiN_x layer rolled the 2D graphene/SWCNTs heterojunction photodetector into a 3D microtubular photodetector.

(j) After being rinsed in deionized water and dried naturally, the 3D graphene/SWCNTs heterojunction photodetector was obtained.

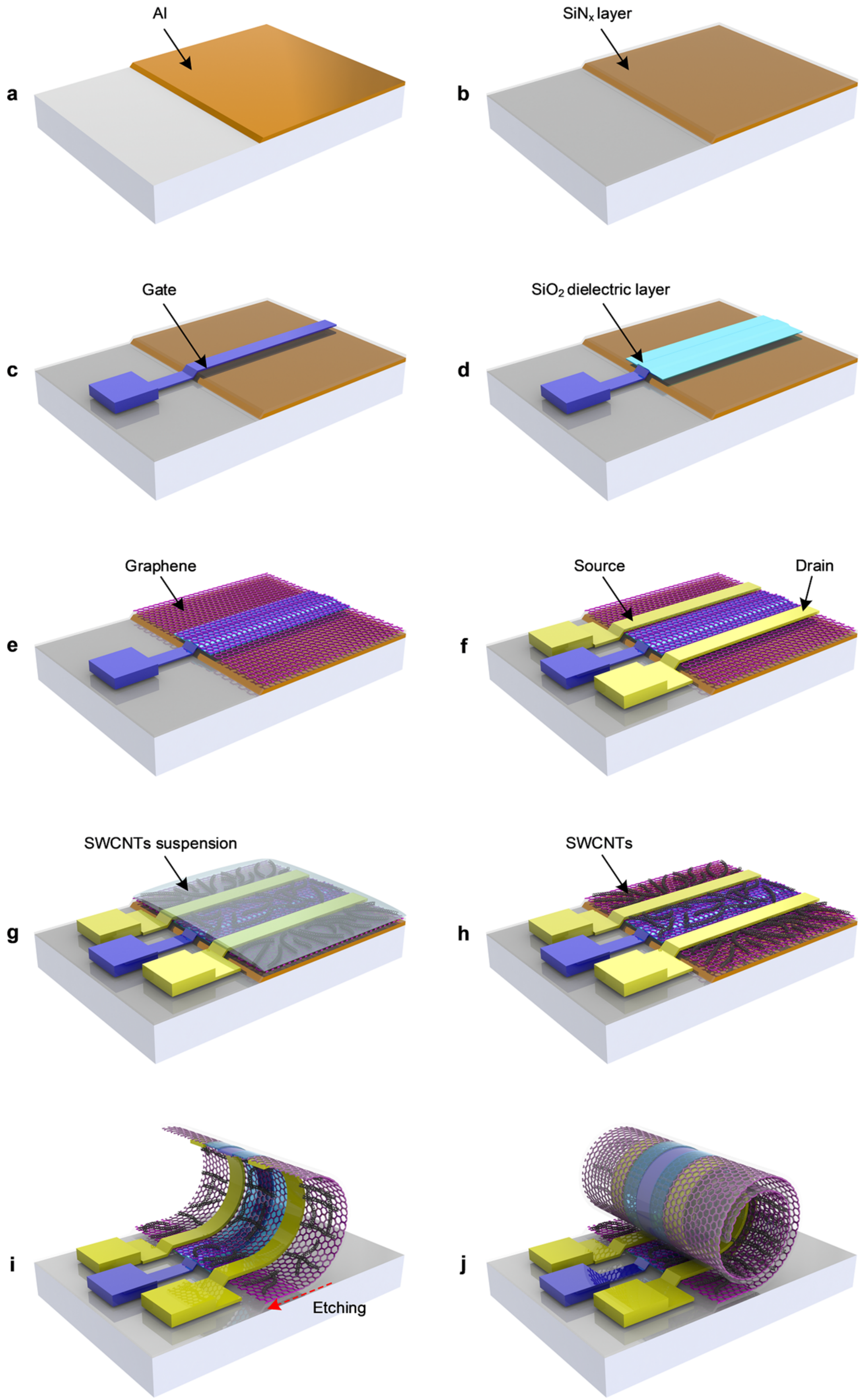

Figure S1. The fabrication process of the 3D microtubular photodetector based on SWCNTs/graphene heterojunction. (a) Clean Si wafer and sputter the Al layer. (b) Deposit SiN_x passivation layer. (c) Sputter Cr/Au buried gate electrode. (d) Deposit SiO_2 dielectric layer. (e) Evaporate Cr/Au source and drain electrodes. (f) Transfer and pattern single-layer graphene. (g) Drop SWCNTs suspension onto 2D GFET. (h) Dry out the 2D GFET with SWCNTs. (i) Etch the Al sacrificial layer selectively. (j) Rinse and dry the 3D SWCNTs/graphene heterojunction photodetector.

2. The resistance profiles of the 2D and 3D heterojunction photodetectors

Figure S2 shows the resistive behavior of the SWCNTs/graphene heterojunction photodetector before (2D FET) and after (3D FET) self-rolled-up process. The carrier mobility (μ) and contact resistance (R_{contact}) of the 2D and 3D FET are analyzed applying a model proposed by Kim *et al.* [1], as shown in below.

$$R_{\text{total}} = R_{\text{contact}} + R_{\text{channel}} = R_{\text{contact}} + \frac{1}{\mu e\sqrt{{n_0}^2 + n^2}}\frac{L}{W}, \tag{1}$$

where R_{channel} is the resistance of the graphene channel, L and W are the length and width of the graphene channel, respectively, e is the electronic charge, n_0 is the density of carriers at the maximum resistance (Dirac point), and n is the gate voltage modulated carrier concentration. By fitting this model to the measured data in Fig. S2, the contact resistance R_{contact} and carrier mobility μ can be extracted. For the 2D heterojunction photodetector (2D FET, Fig. S2a), the $R_{\text{contact,2D}}$ = 307 Ω and μ_{2D} = 1160 $cm^2\ V^{-1}\ s^{-1}$. After rolled into the 3D microtubular photodetector (3D FET, Fig. S2b), the $R_{\text{contact,3D}}$ increases to 332 Ω and μ_{3D} decreased to 1129 $cm^2\ V^{-1}\ s^{-1}$ slightly.

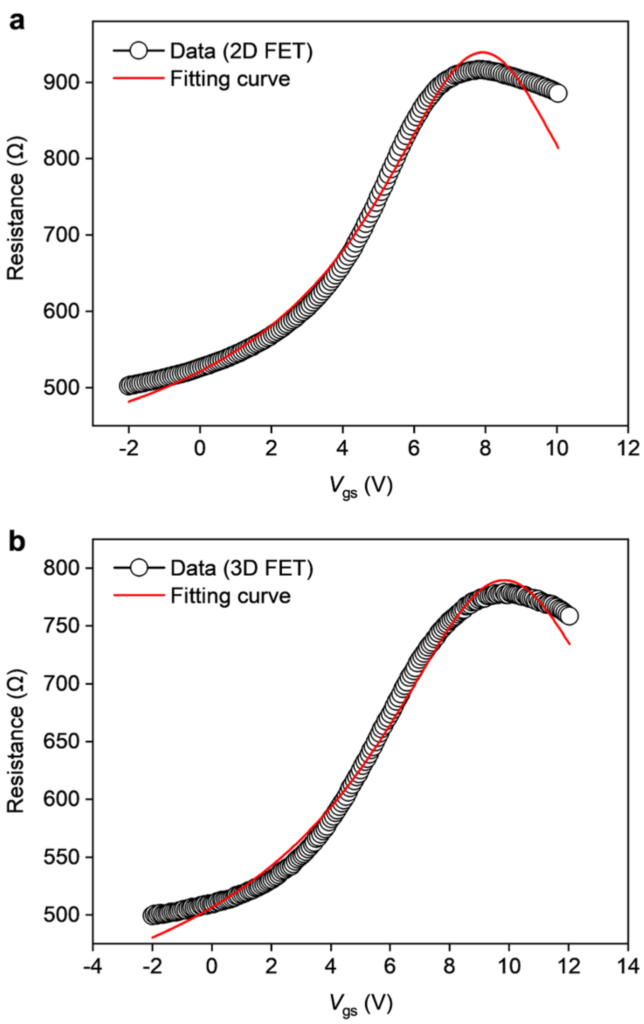

Figure S2. The resistances of the 2D and 3D heterojunction photodetectors. The resistance and fitting results as a function of the gate voltage (V_{gs}) for the heterojunction photodetectors (a) before and (b) after self-rolled-up process.

3. The scanning electron microscopy (SEM) of the 3D heterojunction photodetector

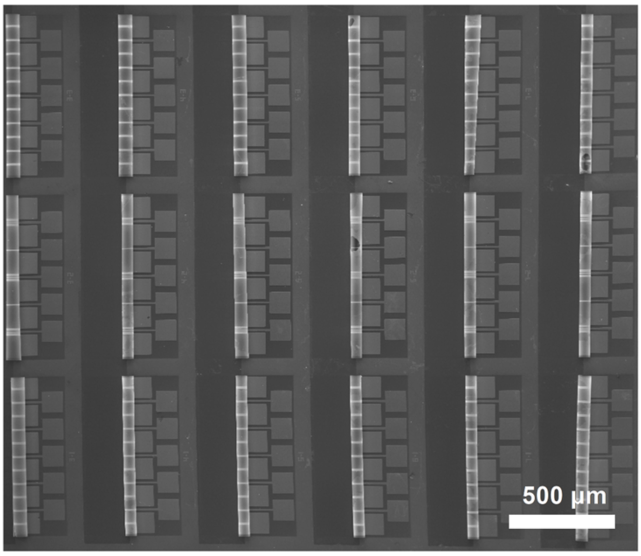

Figure S3. The SEM of 3 × 6 array of 3D microtubular photodetectors.

4. The resistance profile of the 3D heterojunction photodetector applied for photoresponse measurement.

Figure S4 exhibits the resistive behavior of the 3D heterojunction photodetector used for photoresponse measurement. By fitting the curves, the contact resistance R_{contact} and carrier mobility μ can be calculated as 92 Ω and 1665 $cm^2\ V^{-1}\ s^{-1}$, respectively.

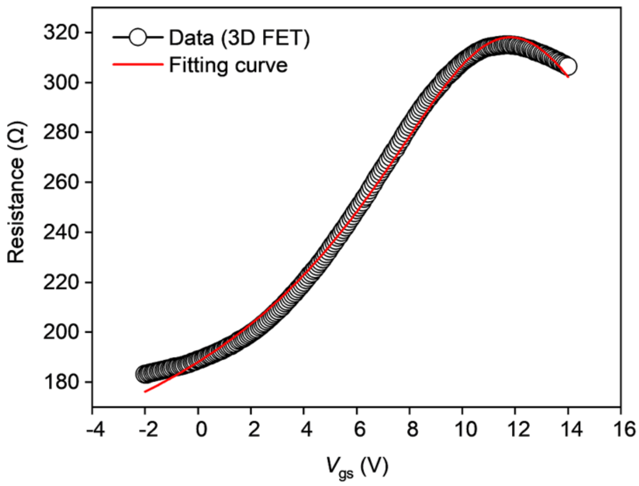

Figure S4. The resistive behavior of the 3D heterojunction photodetector used for photoresponse measurement.

5. The photoresponse of the 2D and 3D heterojunction photodetectors.

Figure S5 depicts the photoresponse of the heterojunction photodetector before (2D FET) and after (3D FET) self-rolled-up process. Under the illumination of 590 nm with the power density of 0.76 mW/cm^2 at V_{gs} = 0 V and V_{ds} = 0.1 V, the photoresponsivity (R_{ph}) of 2D FET is 113 A/W. After etching, the 3D FET achieved a R_{ph} as high as 549 A/W, which is about 5 times higher than that of the 2D FET. This can be contributed to that the 3D natural resonant microcavity enhanced the electric field and increased light-heterojunction interaction area, thus resulting in a high R_{ph} [2].

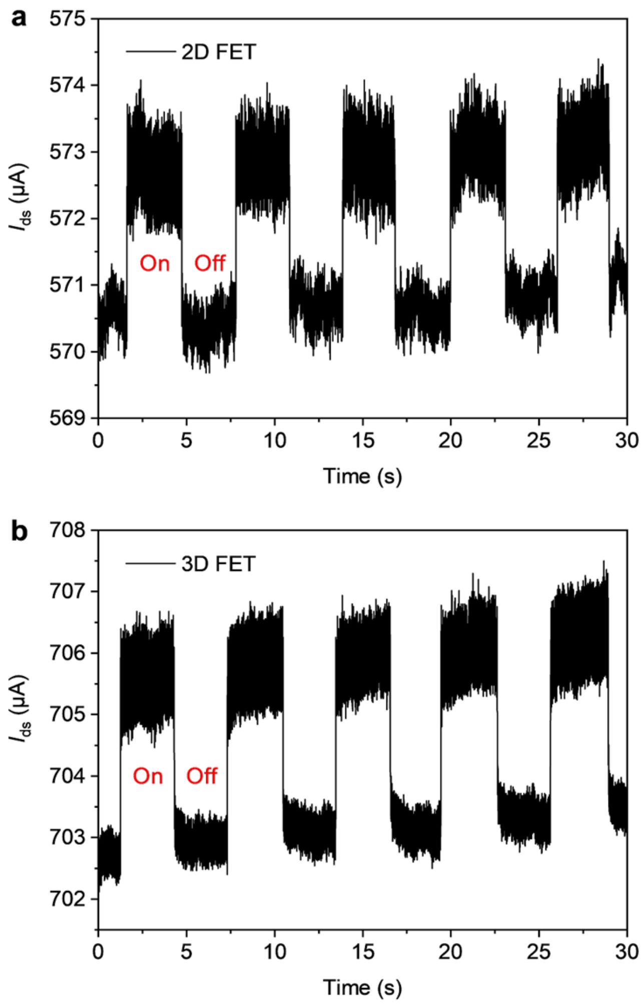

Figure S5. The R_{ph} of the 2D and 3D heterojunction photodetectors. The R_{ph} of the heterojunction photodetectors (a) before (2D FET) and (b) after (3D FET) self-rolled-up process.

The illuminated wavelength is 590 nm with the power density of 0.76 mW/cm^2 at V_{gs} = 0 V and V_{ds} = 0.1 V.

6. The photoresponse of the 3D heterojunction photodetector at 940 nm

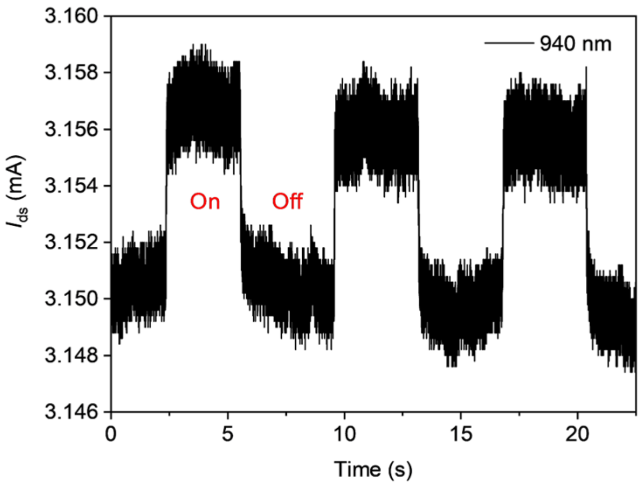

Figure S6. The photoresponse of the 3D heterojunction photodetector. The wavelength of the incident light is 940 nm with the power of 0.3 nW at V_{gs} = 0 V and V_{ds} = 0.5 V. The photoresponsivity can be calculated as 1.9 × 10^4 A/W.

7. The photoresponse of the 3D heterojunction photodetector at 10.6 μm

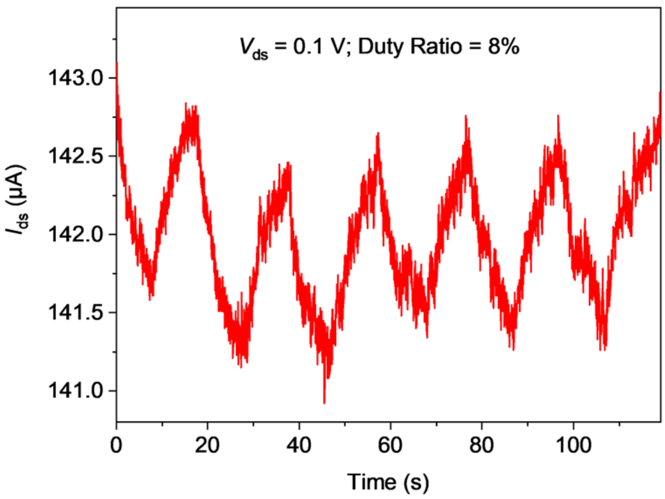

Figure S7. The on−off photocurrent characteristic of the 3D SWCNTs/graphene heterojunction photodetector at 10.6 μm with a duty ratio of 8% when V_{ds} = 0.1 V and V_{gs} = 0 V.

8. The optical scanning system

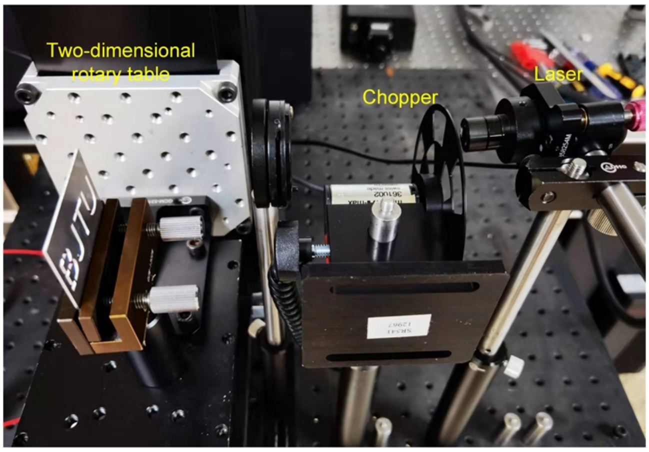

Figure S7. The Optical scanning system for imaging.

References

[1] Kim S, Nah J, Jo I, Shahrjerdi D, Colombo L, Yao Z, Tutuc E and Banerjee S K 2009 Realization of a high mobility dual-gated graphene field-effect transistor with Al_2O_3 dielectric *Appl. Phys. Lett.* 94 062107.

[2] Three-Dimensional Graphene Field-Effect Transistors as High- Performance Photodetectors