nature nanotechnology
SUPPLEMENTARY INFORMATION
DOI: 10.1038/NNANO.2013.219

Highly efficient gate-tunable photocurrent generation in vertical heterostructures of layered materials

Woo Jong Yu, Yuan Liu, Hailong Zhou, Anxiang Yin, Zheng Li, Yu Huang, and Xiangfeng Duan

1. Schematic illustration of the device fabrication process.

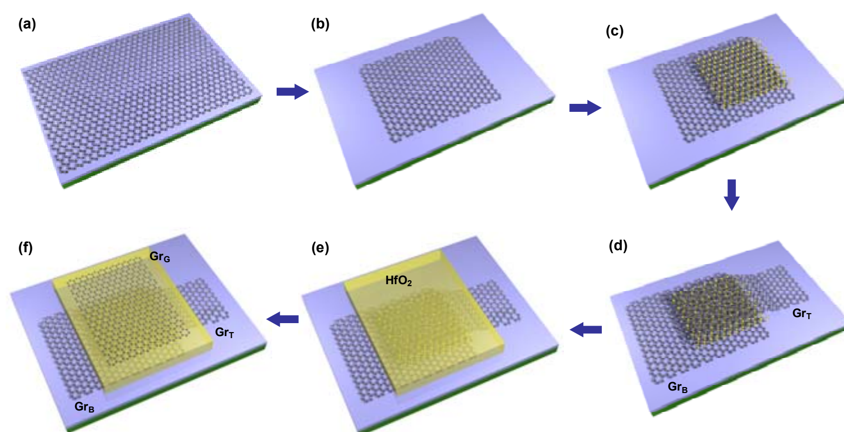

Figure S1l Schematic illustration of the device fabrication procedures. a, CVD grown monolayer graphene was transferred onto a 300-nm SiO_2 covered silicon substrate[1-4]. **b,** Bottom graphene was patterned by oxygen plasma etching using photo resist as a etching mask. **c,** MoS_2 layer was exfoliated onto the graphene through a micromechanical cleavage approach[5]. **d,** The top graphene electrode was transferred and patterned on the MoS_2 to overlap with MoS_2 and bottom graphene. e, For the dual-gate heterostructures, a 60-nm of HfO_2 dielectric layer was deposited by e-beam evaporation, followed by the transferring of another graphene layer as the top-gate electrode.

2. Temporal photoresponse in the vertical graphene-MoS_2-graphene device.

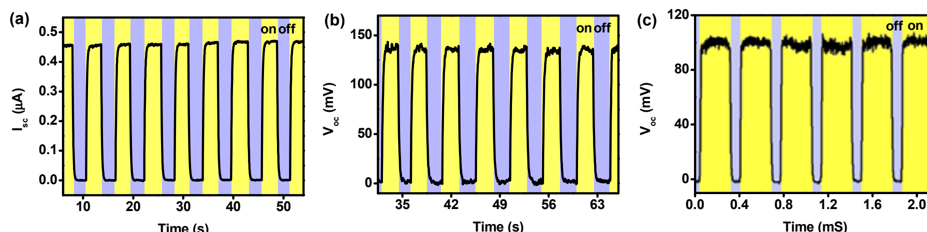

Figure S2l Temporal photoresponse in the vertical graphene-MoS_2-graphene device. a, Time dependent photocurrent response and **b,** Photovoltage response under a global laser (478 nm) illumination with alternatively laser on- and off-periods.. **c,** Time dependent photovoltage measurement under a global laser (478 nm) illumination through a 2.6 kHz chopper. The periodic photoresponse characteristics exhibit the same frequency as that of the laser on-off cycles. The observed photocurrent on-off transition is less than 50 μs, which is only limited by the speed of the mechanical chopper and our measurement capability. The intrinsic speed of the photocurrent generation is likely much higher.

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3. Electrical characteristics of graphene-MoS_2-graphene device.

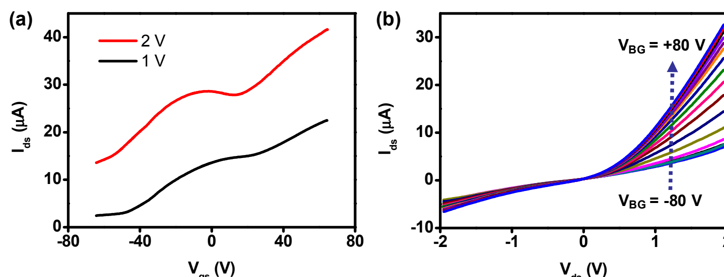

Figure S3l Electrical characteristics of graphene-MoS_2-graphene device. a, Transfer characteristics of graphene-MoS_2-graphene device with silicon substrate as the back-gate at V_{ds} = 1 V and 2 V. **b,** Output characteristics of the same device. The back-gate voltage is varied from −80 V (top) to 80 V (bottom) in the step of 10 V.

4. Simulation of the band diagram of the vertical heterostructure device.

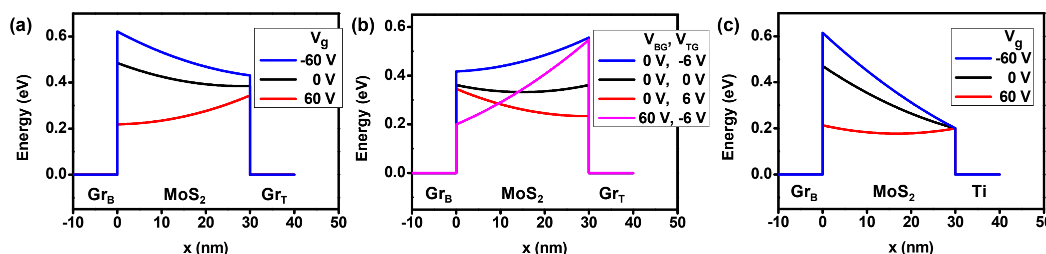

Figure S4l Simulation of the band diagrams. a, Simulated band diagram of single-gated graphene-MoS_2-graphene (GMG) stack. **b,** Simulated band diagram of dual-gated GMG stack. **c,** Simulated band diagram of single-gated graphene-MoS_2-Ti (GMM) stack.

To calculate the band slope and charge distribution, the configurations of GMG and GMM are considered in our simulation. Since MoS_2 is a semiconductor, we adapted the depletion approximation, which means MoS_2 is uniformly charged, and the charge density equals to its doping level. Therefore, bands of MoS_2 are parabolic instead of linear. This model is reasonable when the device channel is short, as in our cases.

For the simulation of GMG, we consider the electric field induced by gate as E_g,

$$E_g = V_g / D \quad (1)$$

where V_g is the gate voltage and D is the thickness of SiO_2 dielectric.

Then we consider electric fields in the two grapheme-MoS_2 interfaces as E_1 and E_2. The carrier density n_1 and n_2 for bottom graphene and top graphene, respectively, can be given by:

$$\varepsilon_l E_g + \varepsilon_s E_1 = n_1 e \quad (2)$$

$$\varepsilon_s E_2 = n_2 e \quad (3)$$

where ε_1 and ε_2 are the dielectric constant for SiO_2 and MoS_2, respectively.

E_1 and E_2 satisfy:

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$$E_1 = E_2 + \frac{Ned}{\varepsilon_s} \qquad (4)$$

where N is the doping level in MoS_2. In our simulation, N is chosen to be 10^{17} cm^{-3}, which can be roughly estimated from transport characteristics of MoS_2 used in our study.

If we have E_1 and E_2, we can get the potential drop ΔV in MoS_2 as:

$$\Delta V = \frac{1}{2}(E_1 + E_2)d \qquad (5)$$

For graphene, we have the relation between carrier density n and chemical potential μ (Dirac point as zero) as:

$$\mu = \frac{h}{2\pi} v_F \sqrt{\pi \,|\, n - n_0 \,|} \qquad (6)$$

where h is the plank constant and v_F is the Fermi velocity, n_0 is the fixed charge graphene. As we know, intrinsic graphene in air is p-doped, and the fix charge can come from SiO_2 substrate or absorbed H_2O and O_2 molecules.

When a bias voltage V_b is applied, we get:

$$eV_b = e\Delta V + \mu_2 - \mu_1 \qquad (7)$$

For GMM, it will be different because the band of MoS_2 are pinned in the metal side. Equation (7) should be replaced by:

$$eV_b = e\Delta V - \mu_1 + W_g - W_m \qquad (8)$$

where W_g is the potential drop from the vacuum level to Dirac point of graphene, W_m is work function of metal. Another difference is that n_2 should be the charge density in the interface of MoS_2 and metal.

Equations above are solved self-consistently to obtain the band diagrams.

5. Simulation of gate dependant depletion width of graphene-MoS_2 schottky contact.

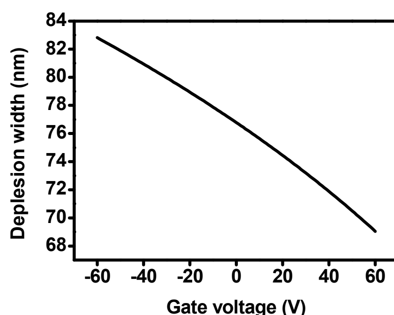

Figure S5I Simulated depletion width in graphene-MoS_2 schottky contact at the various gate voltages. In the graphene-MoS_2-graphene device, the total depletion width at both contact is doubled to be around 140-170 nm.

The electric field induced by gate E_g can be written as the following:

$$E_g = V_g / D \qquad (1)$$

where V_g is the gate voltage and D is the thickness of SiO_2 dielectric.

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Considering the electric field in the graphene-MoS_2 interface as E, the carrier density n for the graphene can be given by:

$$\varepsilon_1\varepsilon_0 E_g + \varepsilon_2\varepsilon_0 E = ne \qquad (2)$$

where ε_1 and ε_2 are the dielectric constant for SiO_2 and MoS_2, respectively.

E satisfies:

$$E = \frac{Ned}{\varepsilon_2\varepsilon_0} \qquad (3)$$

where N is the doping level in MoS_2, and d the depletion length. In our simulation, N is chosen to be 10^{17} cm^{-3}, which can be roughly estimated from transport characteristics of MoS_2 used in our study.

We can get the potential drop ΔV in MoS_2 as:

$$\Delta V = \frac{1}{2} Ed \qquad (4)$$

For graphene, we have the relation between carrier density n and chemical potential μ (Dirac point as zero) as:

$$\mu = \frac{h}{2\pi} v_F \sqrt{\pi |n - n_0|} \qquad (5)$$

where h is the plank constant and v_F is the Fermi velocity, n_0 is the fixed charge graphene. As we know, intrinsic graphene in air is p-doped, and the fix charge can come from SiO_2 substrate or absorbed H_2O and O_2 molecules.

$$eV_b = e\Delta V - \mu + W_g - W_{MoS_2} \qquad (6)$$

where W_g is the potential drop from the vacuum level to Dirac point of graphene, W_m is the work function (4.55 eV)[6] of MoS_2. d is calculated by solving above equations. p-doped graphene (5.0 eV) was used in this calculation.

6. Optical absorption spectroscopy of multi-layer MoS_2 flake.

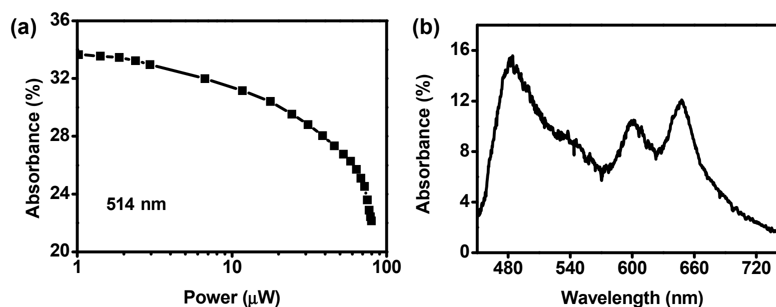

Figure S6I Optical absorption spectroscopy of multi-layer MoS_2 flake. a, Power dependent optical absorption of a multi-layer MoS_2 flake. The absorption was determined by comparing the focused laser (514 nm) transmission through the same glass substrate on and off an MoS_2 flake. The thickness of the MoS_2 flake is about 56 nm. **b,** Absorption spectra of a multi-layer MoS_2 flake on glass substrates. The thickness of MoS_2 flake is about 16 nm. The absorption spectra of MoS_2 was determined from the reflectance measurements[7,8]. To obtain the absorption spectrum, the reflectance spectrum from the MoS_2 flake on a glass substrate (R_{m+s}) and that from

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the same bare glass substrate (R_s) were measured using an optical microscope coupled with a spectrometer and CCD camera. The fractional change in the reflectance, δ_R, can be determined as the difference of these two quantities divided by the reflectance from the bare glass substrate ($\delta_R = \frac{R_{m+s} - R_s}{R_s}$). The absorbance of MoS_2 (A) can then be determined by using the relation: $\delta_R = \frac{4}{n_{sub}^2 - 1} A$, where n_{sub} is the refractive index of the glass substrate.

7. Schematic illustration of the fabrication procedure of graphene-MoS_2-metal (Ti) device.

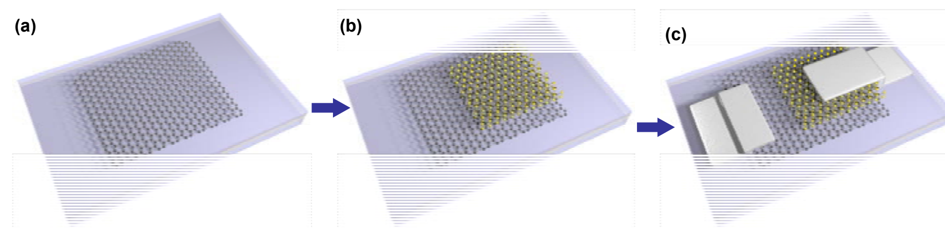

Figure S7I Schematic illustration of the fabrication procedure for graphene-MoS_2-metal (Ti) device. a, CVD grown monolayer graphene was transferred and patterned on the ITO glass with 30-nm Al_2O_3 dielectric layer[1-4]. **b,** MoS_2 layer was exfoliated onto the graphene through a micromechanical cleavage approach[5]. **c,** As a final step, Ti/Au (50 nm/ 50 nm) was patterned as a top electrode and contact electrode for graphene using e-beam lithography.

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