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Supplementary Material

Two-dimensional large-scale bandgap-tunable monolayer MoS2(1-x)Se2x/graphene heterostructures for phototransistors

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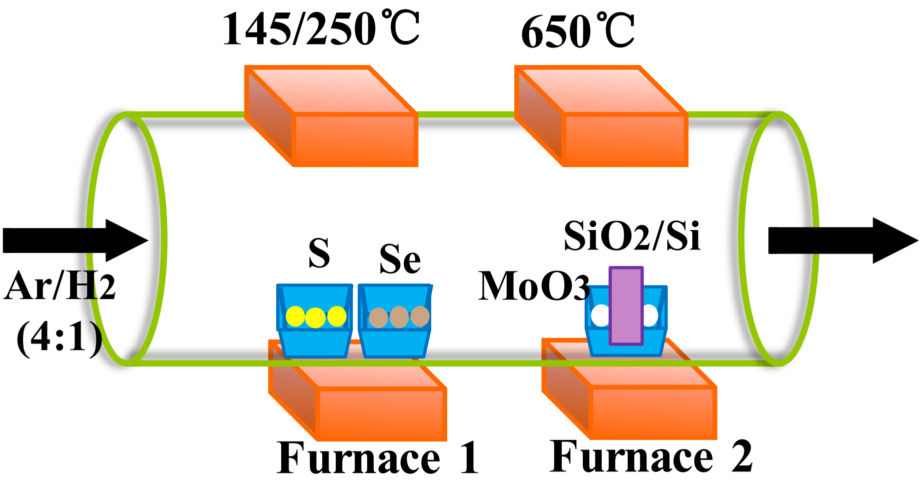

Fig. S1 Schematic illustration for the growth of monolayer $MoS_{2(1-x)}Se_{2x}$ films on the graphene or SiO_2/Si substrate in a tube furnace by LPCVD method.

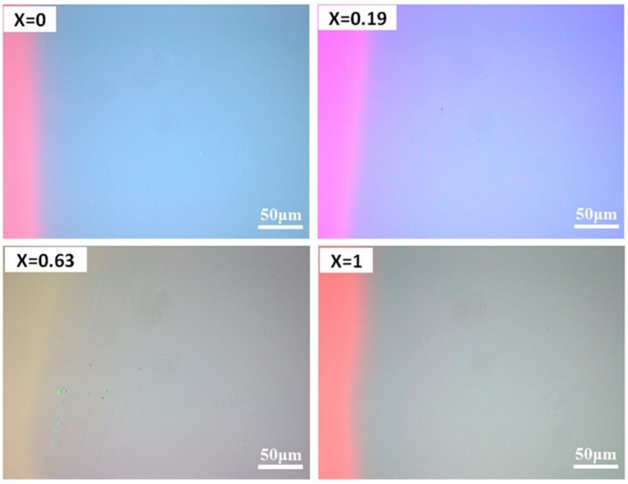

Fig. S2 Optical images of monolayer $MoS_{2(1-x)}Se_{2x}$ with different S composition.

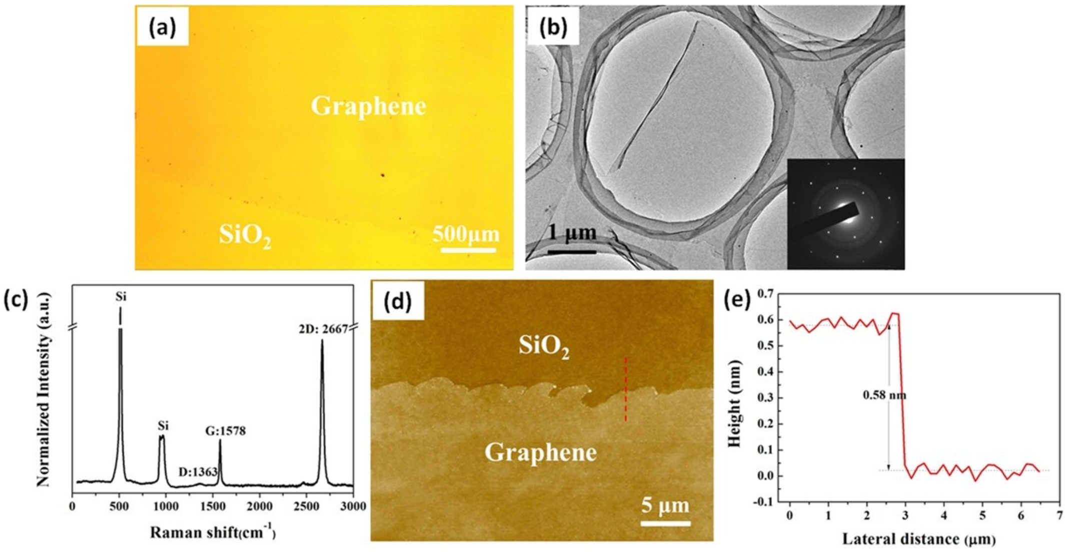

Fig. S3 (a) Optical image of the CVD-grown cm-scale continuous graphene film after transferring to SiO_2/Si substrate. (b) TEM image of the large-scale uniform graphene film covers the carbon frame with the electron diffraction pattern in the inset. (c) Raman spectra of the graphene on SiO_2/Si substrate. (d) AFM image of the graphene film on SiO_2/Si substrate. (e) AFM height profiles of the graphene film from the cross-sectional profile along the red line in (e) indicated the thickness is 0.58 nm.

The large-area graphene film was grown on copper foil using CVD, and then it was transferred on SiO_2/Si substrate. Fig. S3a shows the centimeter-scale continuous and uniform graphene films after transferring to SiO_2/Si substrate. The homogeneous color confirms good thickness uniformity of the film. TEM image in Fig. S2b of the large-scale uniform graphene film covers the carbon frame with the only one set of six fold symmetry diffraction spots in the inset also confirms the single crystalline nature of monolayer graphene film with a hexagonal structure. As indicated by the Raman spectra (Fig. S3c), the extremely low D band suggests high-quality graphene. The AFM image (Fig. S3d, e) shows that the thickness of the film is 0.58 nm, suggesting a monolayer graphene.

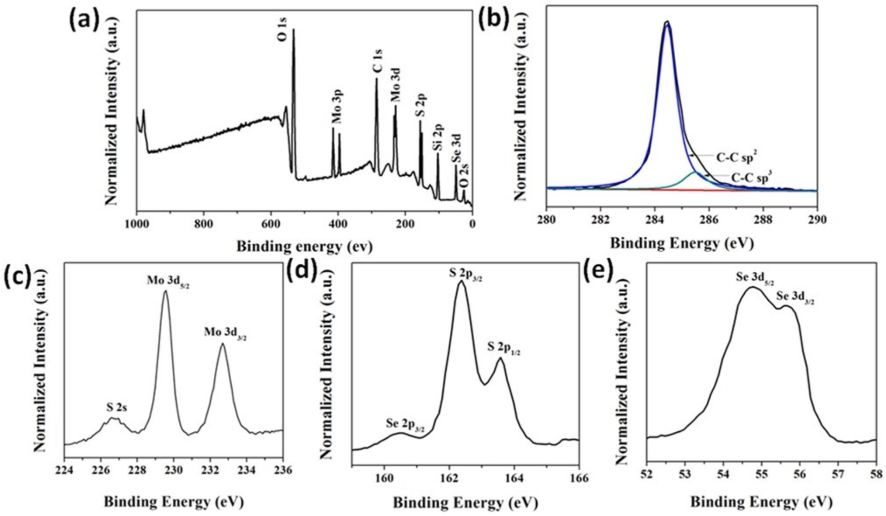

Fig. S4 XPS characterization of the CVD-grown $MoS_{2(1-x)}Se_{2x}$/graphene hetero-structure with S composition of x = 0.52. (a) XPS spectrum of the sample. Six elements are present: Mo, S, Se and C (from the sample), Si and O (from the substrate). (b) XPS spectrum of C 1s. (c) XPS spectrum of Mo 3d (d) XPS spectrum of S 2p. (e) XPS spectrum of Se 3d, Curve fitting is done on the C 1s spectrum to show the dominating sp2 than sp3 bonding of C atoms.

Fig. S4a shows the XPS spectra of the $MoS_{0.96}Se_{1.04}$/graphene heterostructure. The curves in Fig. S4c,d,e are in accordance with the $MoS_{0.96}Se_{1.04}$ film on the SiO_2/Si substrate (Fig. 3). Fig. S4b shows C 1s curve. After curve fitting the signal, it exhibits the sp^2 bonding and sp^3 bonding located 284.48 and 285.48 eV, which are underlying the graphene film. However, we cannot find any peak at near 286.8 eV where the C-S binding energy in the C 1s spectrum. This result indicates that the $MoS_{0.96}Se_{1.04}$ and graphene layers are stacked by the van der Waals force rather than the C-S bond.

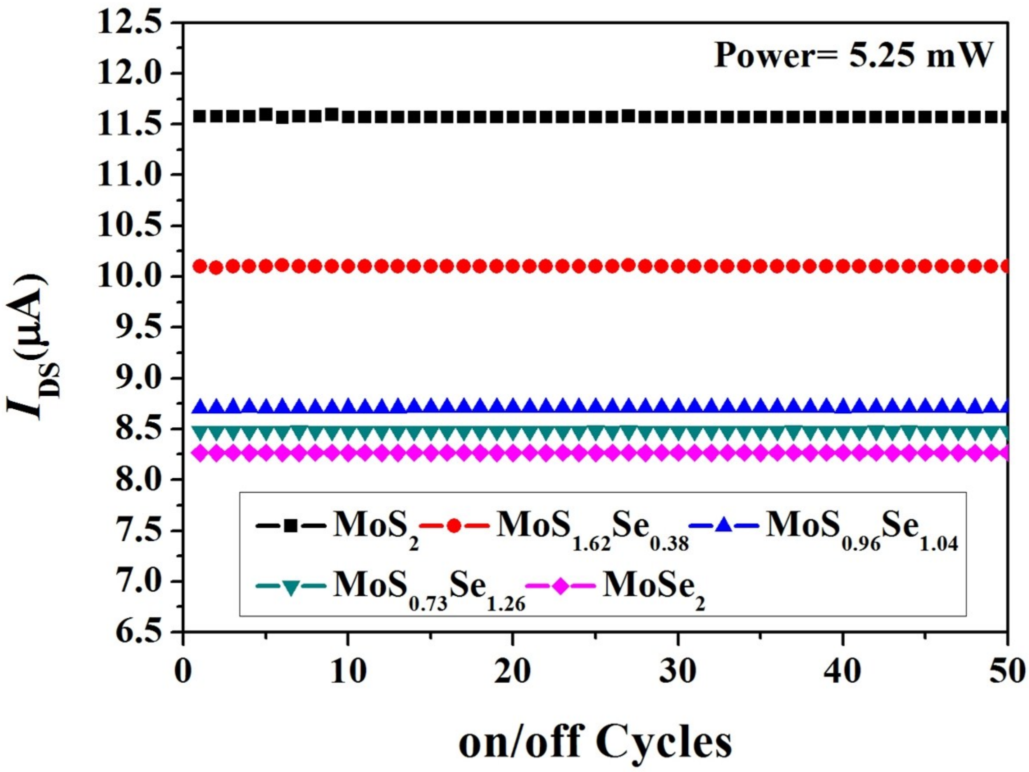

Fig. S5 I_{DS} of a photoswitch during 50 cycles for phototransistors of $MoS_{2(1-x)}Se_{2x}$/ graphene heterostructure under light irradiation of 5.25 mW (V_{DS} = 0.01 V).

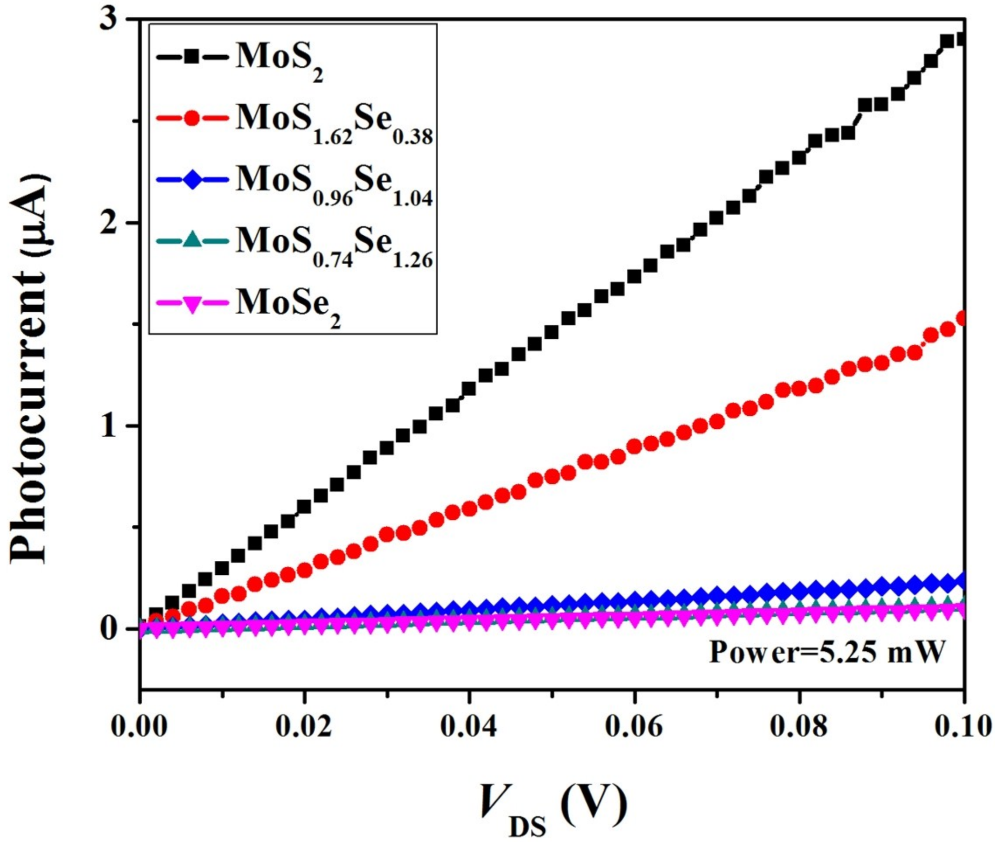

Fig. S6 Photocurrent of the $MoS_{2(1\text{-}x)}Se_{2x}$ /graphene transistor for different Se content showing a linear dependence on the bias voltage (V_{BG} = 0V).

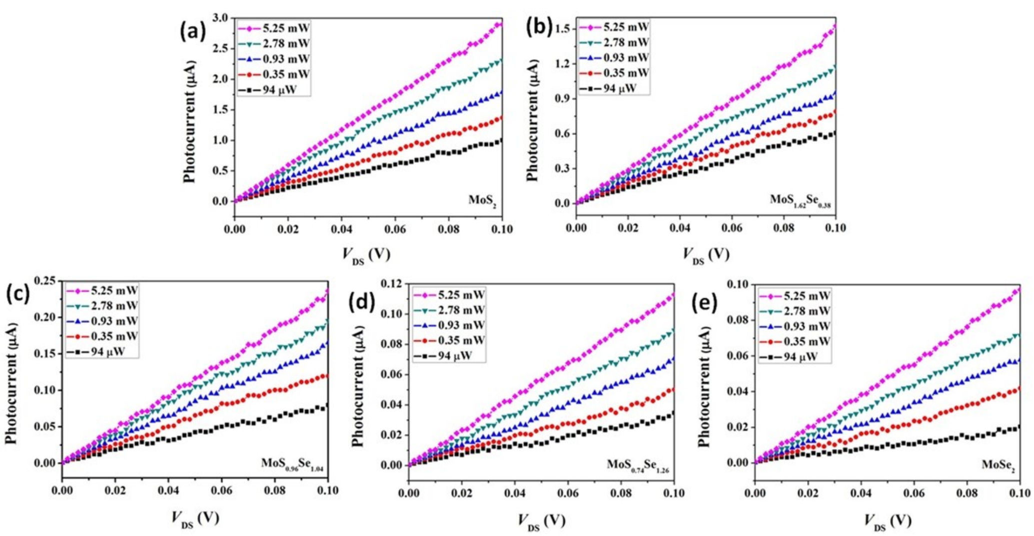

Fig. S7 Photocurrent characteristics of the centimeter-scale $MoS_{2(1-x)}Se_{2x}$/graphene heterostructure ranging in composition from (a) MoS_2 (1.82eV) to (e) $MoSe_2$ (1.66 eV)for different optical power as a function of drain-source voltage (V_{DS}), showing a linear dependence on the bias voltage (V_{BG} = 0).

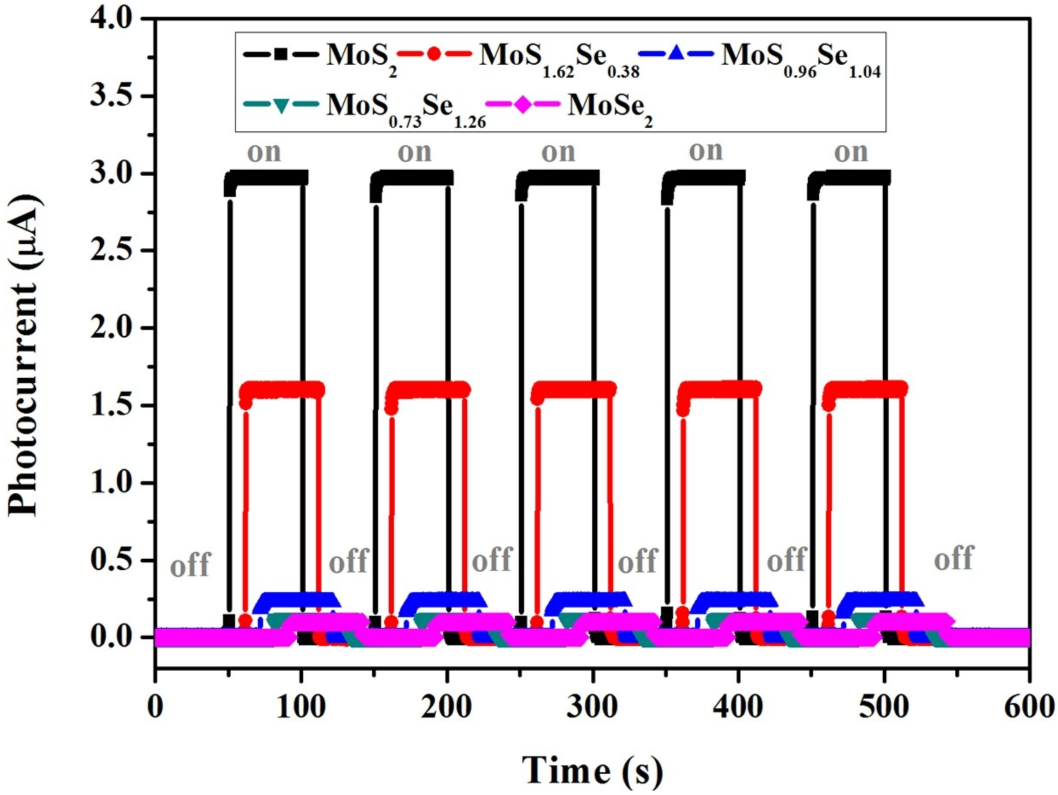

Fig. S8 The of Photocurrent of time-resolved photo-response of the hybrid phototransistor with an on/off switching light of 5.25 mW (V_{DS} = 0.01 V, V_{BG} = 0V).

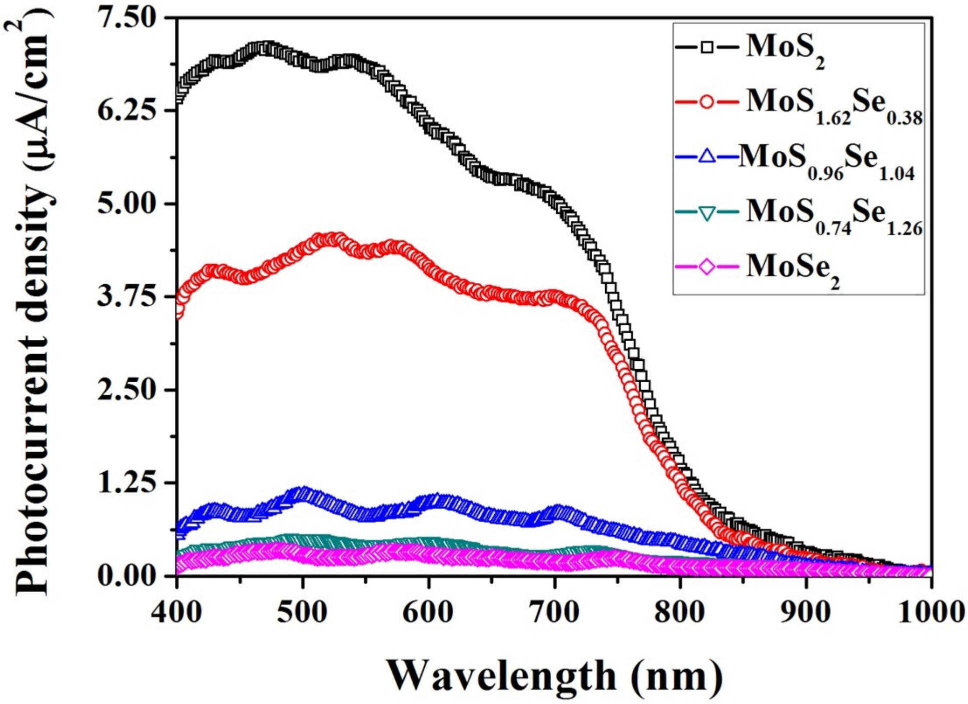

Fig. S9 The Photocurrent response of the $MoS_{2(1-x)}Se_{2x}$/graphene heterostructure phototransistor as a function of the wavelength of irradiation light (V_{DS} = 0.01 V, V_{BG} = 0V).

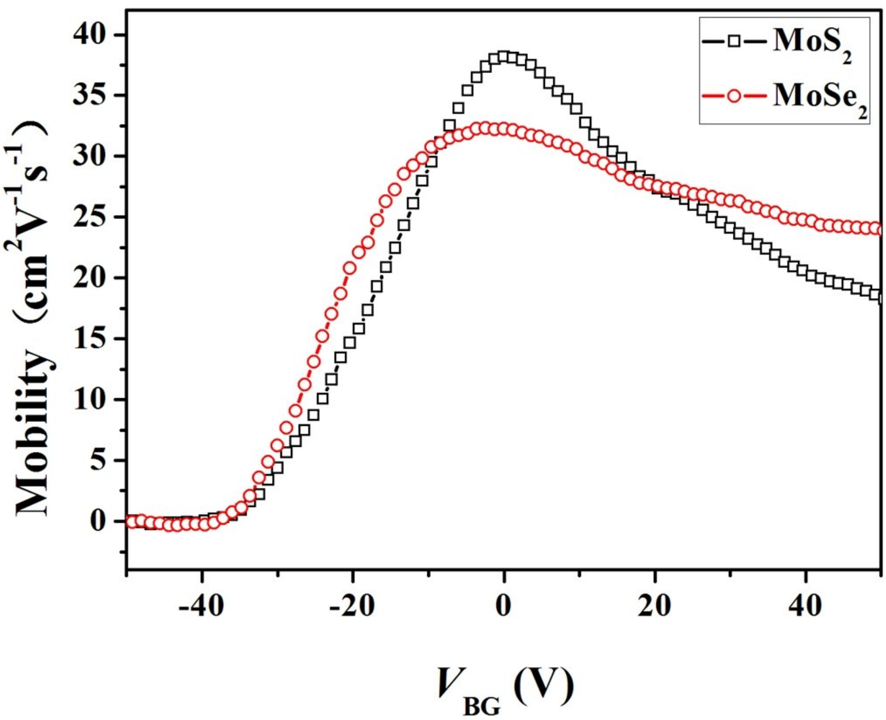

Fig. S10 Field effect mobilities of the MoS_2 and $MoSe_2$ films extract from our heterostructure phototransistors under light irradiation (V_{DS} = 0.01 V).

The field effect mobility is calculated using expression $\mu=[dI_d/dV_{bg}]\times[L/(WC_{ox}V_d)]$, where V_{bg} and V_d are the back-gate voltage and source-drain voltage, L and W are the channel length and width, and C_{ox} is the capacitance per unit area between the channel and the back gate (calculated from 300 nm SiO_2). The Field effect mobilities of the MoS_2 and $MoSe_2$ films reach the peak value of 38.2 $cm^2/(V\ s)$ and 32.2 $cm^2/(V\ s)$ respectively.

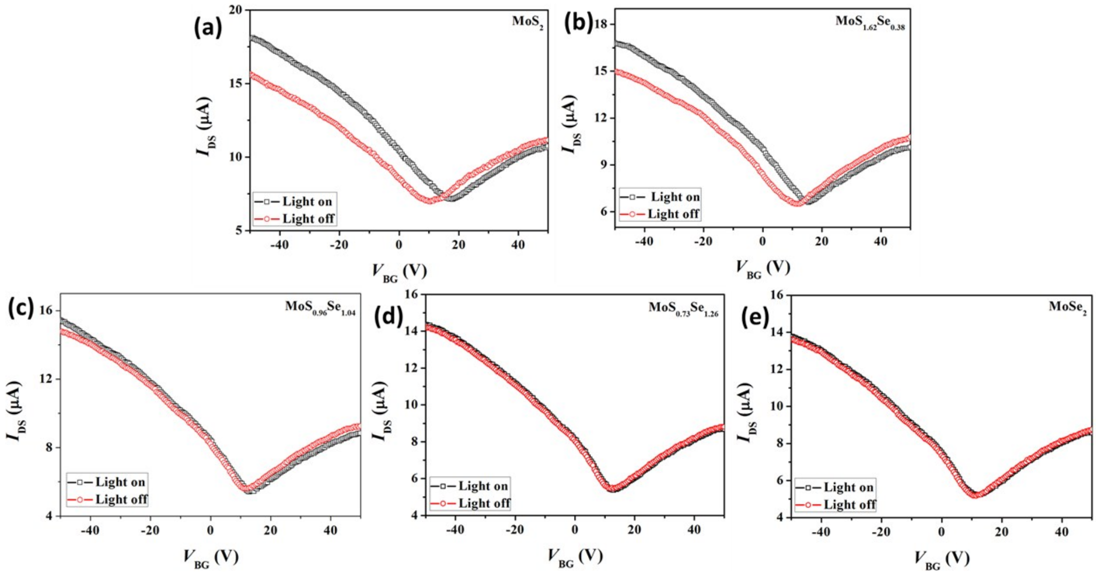

Fig. S11 Representative device characteristics of the $MoS_{2(1-x)}Se_{2x}$/graphene heterostructure phototransistor under light irradiation and in the dark (V_{DS} = 0.01 V).

Transfer of $MoS_{2(1-x)}Se_{2x}$ film Cu TEM grids

The as-synthesized $MoS_{2(1-x)}Se_{2x}$ films were transferred from the growth substrate onto Cu TEM grids using the wet transfer NaOH method. The $MoS_{2(1-x)}Se_{2x}$ film was spin coated with a layer of polymethyl methacrylate (PMMA) (Micro Chem. 950K A4) by spin-coating (step 1: 500 rpm for 20 s; step 2: 3000 rpm for 1 min), followed by baking at 150 °C for 10 min. After that, the PMMA-supported $MoS_{2(1-x)}Se_{2x}$ film was then soaked with a NaOH (2 M) solution at 80 °C for 30 min. The NaOH solution etched the SiO_2 layer, causing the PMMA-supported monolayer $MoS_{2(1-x)}Se_{2x}$ film to float on its surface. The PMMA-supported $MoS_{2(1-x)}Se_{2x}$ film was then transferred to deionized (DI) water and washed several times to remove the etchant and residues. Copper grid (for TEM observations) was then used to "fish out" the PMMA-supported $MoS_{2(1-x)}Se_{2x}$ film, which was dried on a hot plate at 100 °C for 10 min to promote the interaction between film and the substrate. The PMMA film was removed by washing sequentially with acetone, isopropyl alcohol (IPA), and finally, DI water.

The quantitative analysis of the ratio of S to Se by XPS

The ratio of S to Se can be calculated from the XPS results by the following formula:

$$S/Se = (I_S \times F_{Se})/(I_{Se} \times F_S)$$

where I_S and I_{Se} are the areas under the S-2$p_{3/2}$ and Se-3$p_{3/2}$ peaks, respectively, *and* F_S and F_{Se} represent the relative symmetric factors (R.S.F) for S-2$p_{3/2}$ (0.4453) and Se-3$p_{3/2}$ (0.8493) respectively.