Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

Highly efficient gas molecule-tunable few-layer GaSe phototransistors

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Experimental section

Mechanical Exfoliation of GaSe: GaSe crystals were grown by modified Bridgman technique. Few-layer GaSe nanosheets were exfoliated from bulk GaSe crystals and then deposited onto the freshly cleaned 300 nm SiO_2/Si substrates using the scotch tape-based micromechanical exfoliation method. We observed few-layer GaSe nanosheets through an optical microscope (OLYMPUS OLS40-SU).

Thermal Annealing: The samples were heated to 300 °C in a 25 °C/min rate and the temperature was held at 300 °C for half an hour in vacuum. After annealing, the furnace was cooled down to room temperature and the samples were taken out of the furnace.

Phototransistors were fabricated as follows: Metal electrodes were fabricated using UV photolithography (SUSS MA6/BA6) followed by electron beam evaporation (EB700-I) of 8 nm Cr and 50 nm gold. Electrical characterization for the phototransistors was measured using a semiconductor parameter analyzer (Agilent B2902A) and shielded probe station.

Raman Spectroscopy: Analysis of the few-layer GaSe nanosheet by Raman spectroscopy was carried out on a laser Raman spectrometer (Renishaw inVia plus) with the excitation line of 514 nm used in a backscattering configuration. Raman spectra were collected using a 100× objective and recorded with 1800 lines/mm grating providing the spectral resolution of ~ 1 cm^{-1}. The spectra were entirely recorded at low power levels P =0.5 mW to avoid laser-induced heating and ablation of the samples. And the photoluminescence (PL) measurements have been carried out with the same Renishaw inVia plus setup.

EDX analysis: Energy dispersive X-ray spectroscopy (EDX) was utilized for the estimation of the composition of GaSe and it was equipped on the SEM instrument (FEI NOVA NanoSEM430).

Density Functional Theory Calculations: Our first-principles spin-polarized calculations are performed based on density-functional theory (DFT) by using the Vienna *ab initio* simulation package (VASP)[1]. A 3×3 supercell of GaSe monolayer is used to simulate the adsorption of O_2 molecule on it. The exchange-correction interaction is treated by the van der Waals density functional (vdW-DF)[2] to describe this adsorbed system. After energy convergence analysis, an energy cutoff of 450 eV for the plane-wave basis set and a Monkhorst-Pack grid of 6×6×1 for the Brillouin zone integration are employed. In order to avoid the interaction between two adjacent monolayers, a vacuum layer larger than 15 Å is added. The geometric structure is fully relaxed by using conjugate gradient method until the Hellmann-Feynman force on each atom is smaller than 0.02 eV/Å. The charge transfer between GaSe monolayer and O_2 molecule is obtained based on the Bader analysis[3].

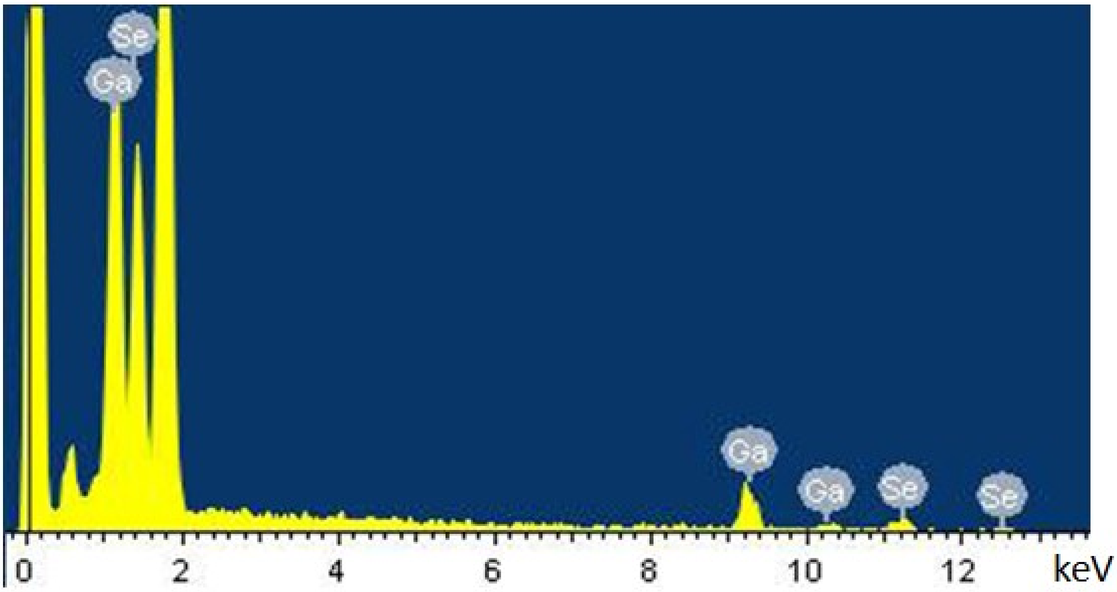

Figure S1. EDX pattern of GaSe nanosheet.

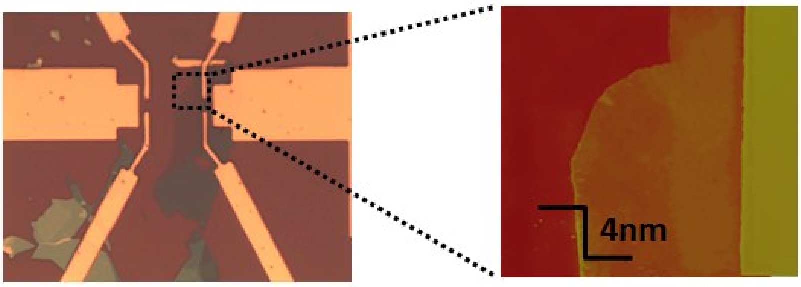

Figure S2. The AFM image of few-layer GaSe nanosheet.

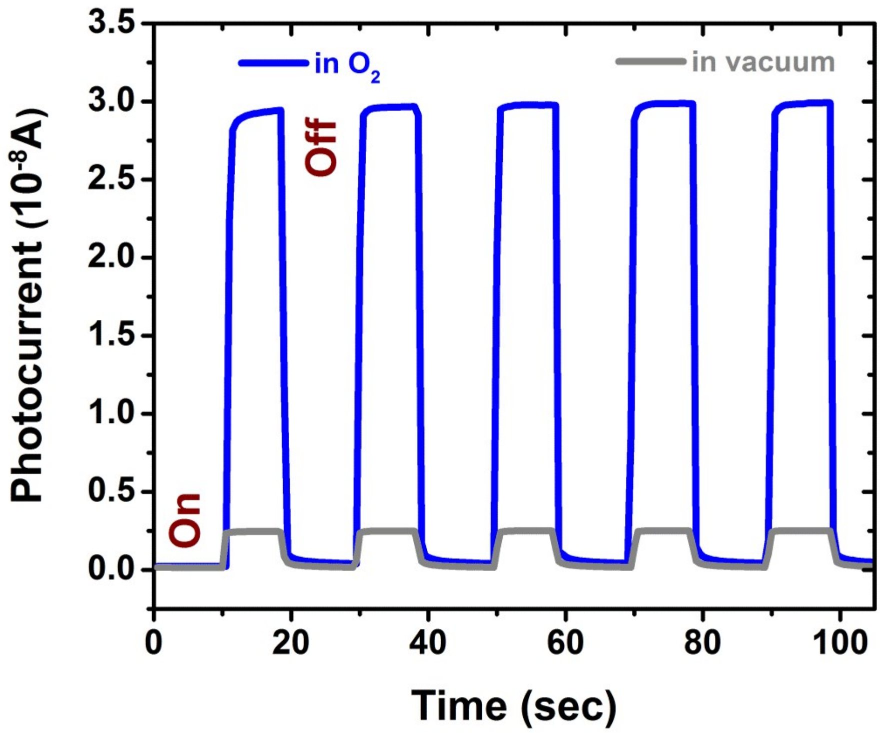

Figure S3. I-t curves of the annealed device in O_2 gas environment (blue) and in vacuum (gray) illuminated by UV light switching on/off.

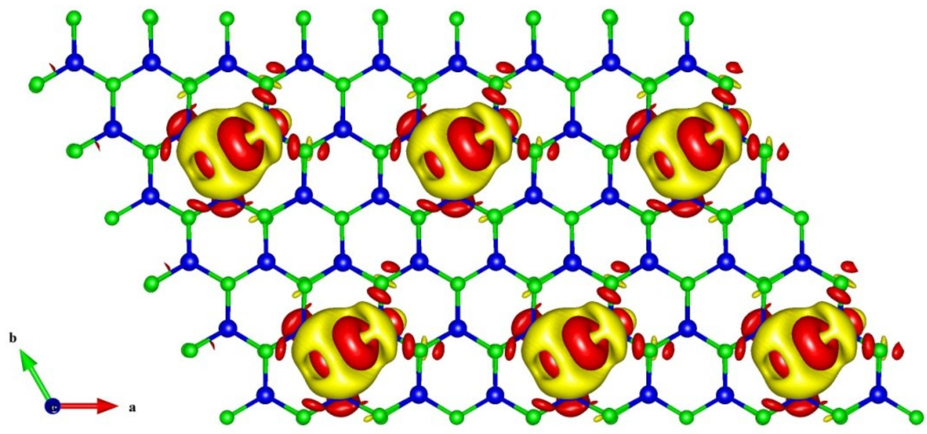

Figure S4. Charge density difference plots for GaSe with defect (top view). The iso-surface value with defect is 1×10^{-3} e/Å^3. Red and yellow distributions correspond to charge accumulation and depletion, respectively.

Table 1 Comparison of the parameters of our device to the reported 2D material based photodetectors.

Photodetectors	**R_λ (AW^{-1})**	**EQE (%)**	**Response time**
Graphene [4]	1.0×10^{-3}	6-16	
Graphene [5]	6.1×10^{-3}	1500	
GaS Nanosheet [6]	4.2	2050	~30 ms
Our phototransistor	18.75	9153	~0.21s

Reference

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