## Highly Responsive Ultrathin GaS Nanosheet Photodetectors on Rigid and Flexible Substrates

PingAn Hu<sup>1</sup>\*, Lifeng Wang<sup>1</sup>, Mina Yoon<sup>2</sup>, Jia Zhang<sup>1</sup>, Wei Feng<sup>1</sup>, Xiaona Wang<sup>1</sup>, Zhenzhong Wen<sup>1</sup>, Juan Carlos Idrobo<sup>3</sup>, Yoshiyuki Miyamoto<sup>4</sup>, David B. Geohegan<sup>2</sup>, Kai Xiao<sup>2</sup>\*

## AUTHOR ADDRESS

<sup>1</sup>Key Lab of Microsystem and Microstructure, Harbin Institute of Technology, Ministry of Education, No. 2 YiKuang Street, Harbin, 150080, P.R. China E-mail: hupa@hit.edu.cn

<sup>2</sup> Center for Nanophase Materials Sciences,<sup>3</sup> Materials Science and Technology Division, Oak Ridge National Laboratory, One Bethel Valley Road, Oak Ridge, TN, 37831, E-mail: xiaok@ornl.gov

<sup>4</sup> Nanosystem Research Institute, National Institute of Advanced Industrial Science and Technology (AIST), Central 2, 1-1-1 Umezono, Tsukuba 305-8568, Japan

KEYWORDS Gallium Sulfide, two-dimensional materials, nanosheets, photodetectors, photoresponsivity

## Supporting Information

The GaS laminar precursor was obtained by the following method: The method uses a two-zone horizontal furnace with a fused silica tube, into which the sulfur powder and gallium were placed in the low temperature zone  $(300! \sim 400!)$  and the high temperature zone  $(850! \sim 910!)$ , respectively. Evaporated sulfur was carried to the high temperature zone by flowing Ar gas (50 sccm) to react with the gallium to form a GaS crystal. The reaction was usually carried out for 20 minutes. The few layer GaS nanoheets were made from a bulk layer precursor by a solvent or mechanical cleavage method. The thickness and layer number were determined by using a Bruker Dimension Icon Atomic Force Microscope (AFM). The composition and structure of the few-layer GaS nanosheets were characterized and analyzed by scanning electron microscopy (SEM, JSM-6301F) equipped with energy dispersion X-ray spectroscopy (EDS), and transmission electron microscopy (TEM, JEOL-2010F). The Z-contrast images were obtained on an aberration-corrected Nion UltraSTEM operating at 100 kV, using a half-angle range of the annular dark field detector of 86 to 200 mrad. Raman spectra were collected for bulk samples and nanosheets of GaS using a Horiba-Jobin Yvon HR800 which is equipped with a liquid-nitrogen-cooled CCD, using 532 nm-laser excitation from a diode pumped solid state laser. The Raman spectroscopy measurements were performed in a backscatter geometry using a 100X objective lens (NA = 0.90) and a 600 or 1800 groove/mm grating (dependent on the spectral resolution) at room temperature. The ultrathin GaS was transferred to a silicon wafer with an oxidized layer of 300 nm. Cr/Au electrodes with 10 nm thick Cr and 100 nm thick Au were fabricated using a shadow mask. The devices with a gap of 20 µm were then annealed at 200 °C to reduce resistance. The light source for the photoresponse data was a 500 W xenon lamp. Optical bandpass filters were used to narrow wavelength regions between 254-610 nm. To

explore the photoresponse behavior of ultrathin GaS nanosheet devices, this narrow band illumination was directed vertically onto the whole device. Photocurrent measurements were carried out using a Lakeshore probe station and an HP 4140B semiconductor parameter analyzer. The intensities of the incident beams were measured by a power meter (Scientech 372). Infrared light was filtered throughout the experiments with a Toshiba IRA-25S IR-cut filter (Japan) to protect the electrodes from heat.

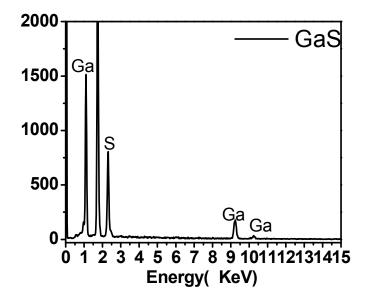


Figure S1 Energy-dispersive X-ray spectroscopy (EDS) of GaS nanosheets

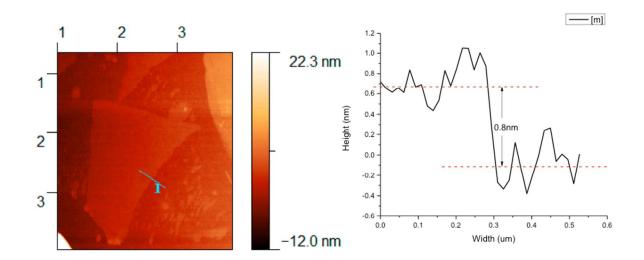


Figure S2 AFM image of single-layer GaS nanosheet and the measured thickness.

Figure S3 GaS device photocurrent as a function of the irradiation time in air. The photocurrent is almost invariant during irradiation times of < 30 min, but significantly decreases during a long-term operation (~40% decrease after 2 hours), most likely possibly due to oxidation in air induced by deep-UV irradiation.

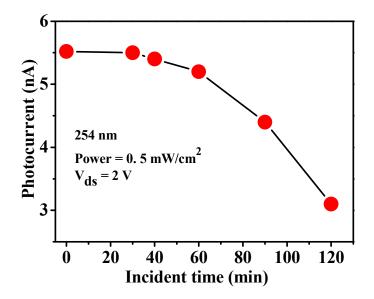


Figure S4 Photocurrent and responsivity measured during bending, measured under 254-nm irradiation at an initial irradiance of 256 mW/cm<sup>2</sup> and  $V_{DS}$ = 2V. The photocurrent and responsivity both decrease with smaller bending angles, e.g. Responsivity is about 7.5 A/W without bending and about 3.6 A/W after bending to 60°. A factor of two decrease in both is expected due to the decrease in irradiance from geometric effects

