Supplementary Information for

A high-performance visible-light-driven all-optical switch enabled by ultra-thin gallium sulfide

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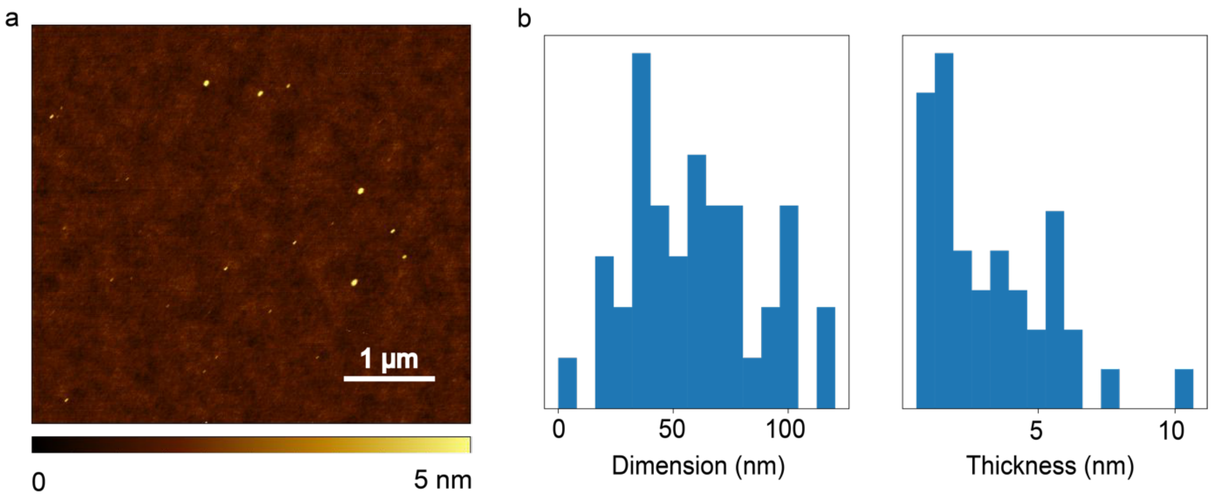

Fig. S1 Statistical analysis of the obtained Ga_2S_3 nanoflakes based on the AFM profiles. a) Overall AFM images. b) Statistical distribution of the lateral dimension and thickness of Ga_2S_3 nanoflakes.

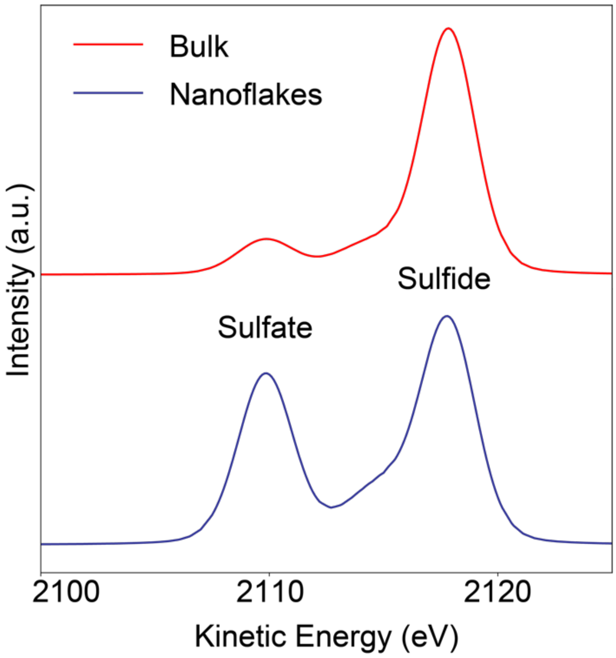

Fig. S2 The S KLL spectrum of Ga_2S_3 for bulk and nanoflakes, in which the sulfate peaks are observed on both samples.

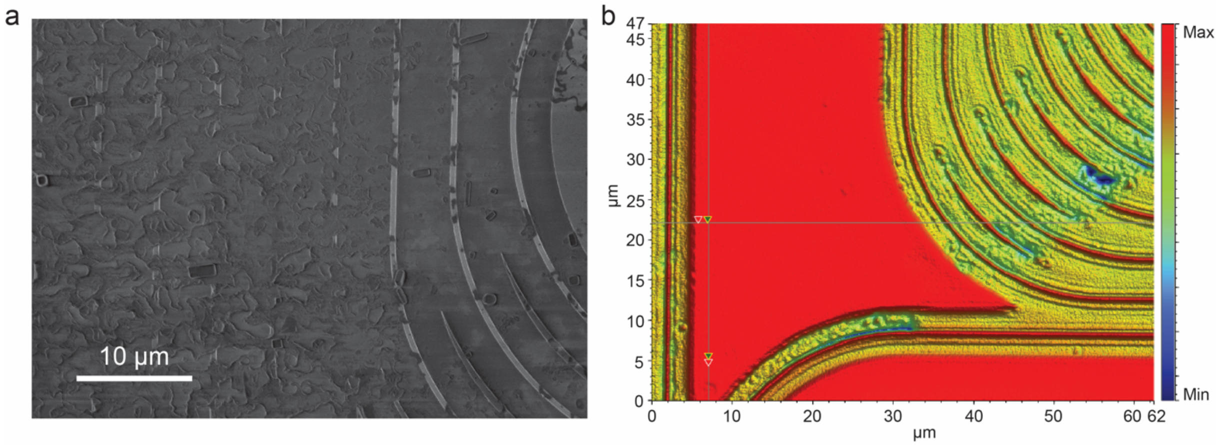

Fig. S3 a) The SEM microscopy image of the device incorporated with the Ga_2S_3 layer. b) The corresponding 3D optical surface profiler image.

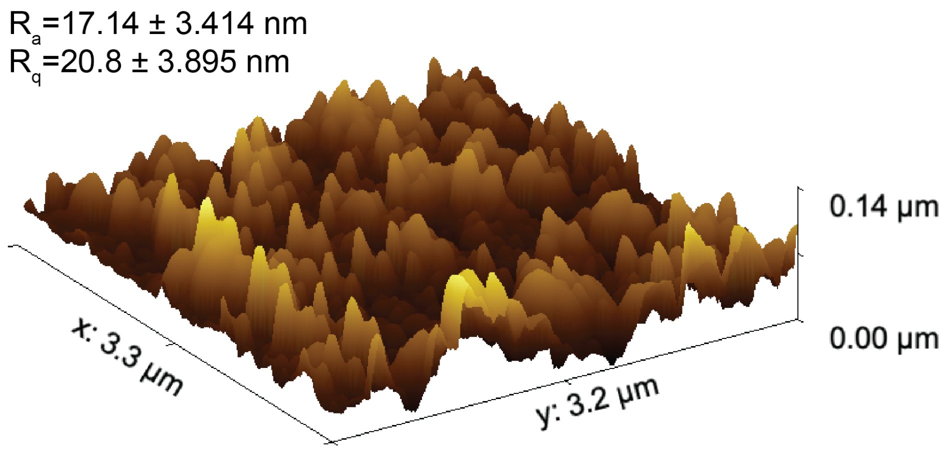

Fig. S4 The surface roughness of the deposited Ga_2S_3 layer extracted from the AFM image.

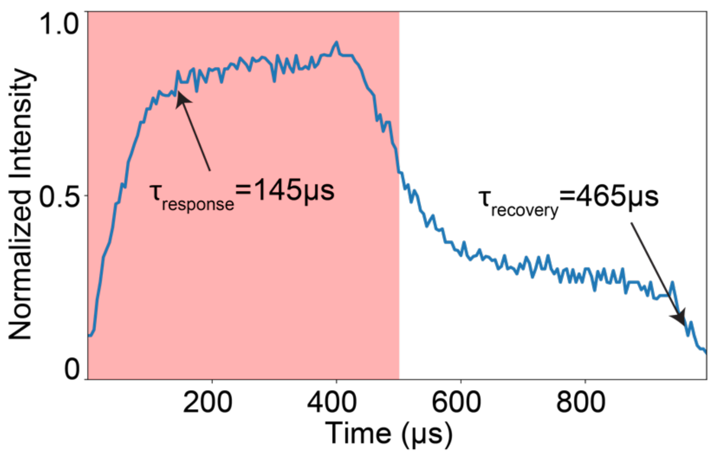

Fig. S5 The 10% to 90% of the response/recovery of the Ga_2S_3 based optic-switch under a 1 kHz of control pulse at the excition wavelength of 532 nm.

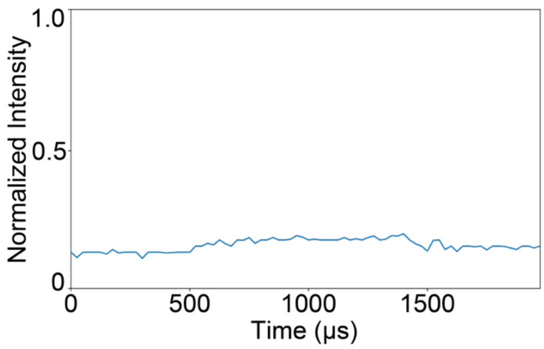

Fig. S6 The switching response of the pure silicon waveguide with 1 kHz of the control pulse at the excitation wavelength of 532 nm.

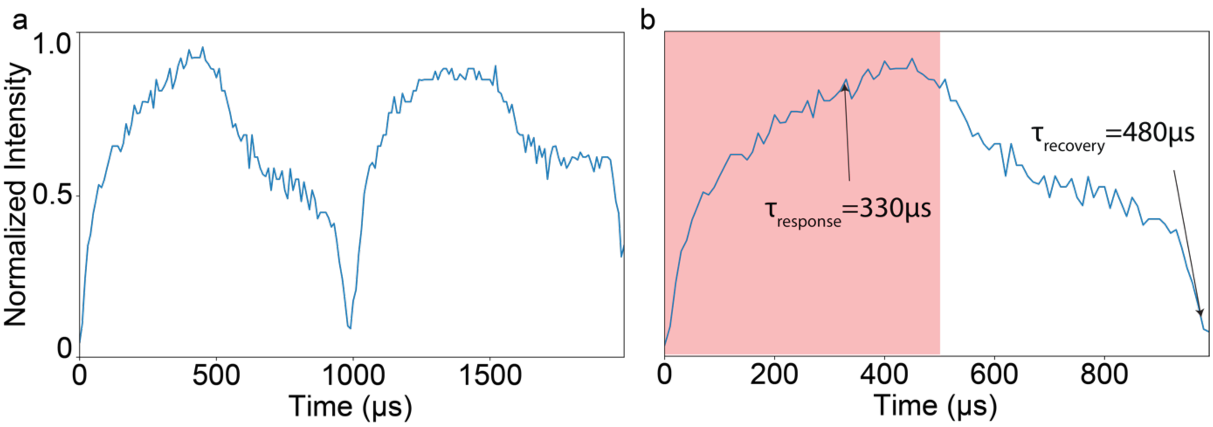

Fig. S7 a) The switching response of Ga_2S_3 based optic-switch with 1 kHz of control pulse at the wavelength of 473 nm. b) The corresponding 10% to 90% of the response/recovery time of the Ga_2S_3 optic-switch.

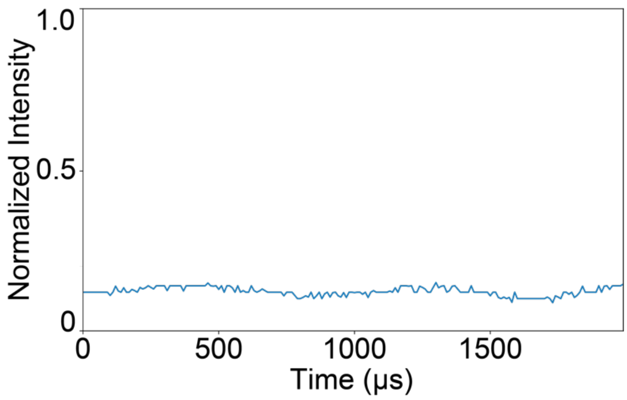

Fig. S8 The switching response of Ga_2S_3 based optic-switch with 1 kHz of control pulse at the wavelength of 632 nm.

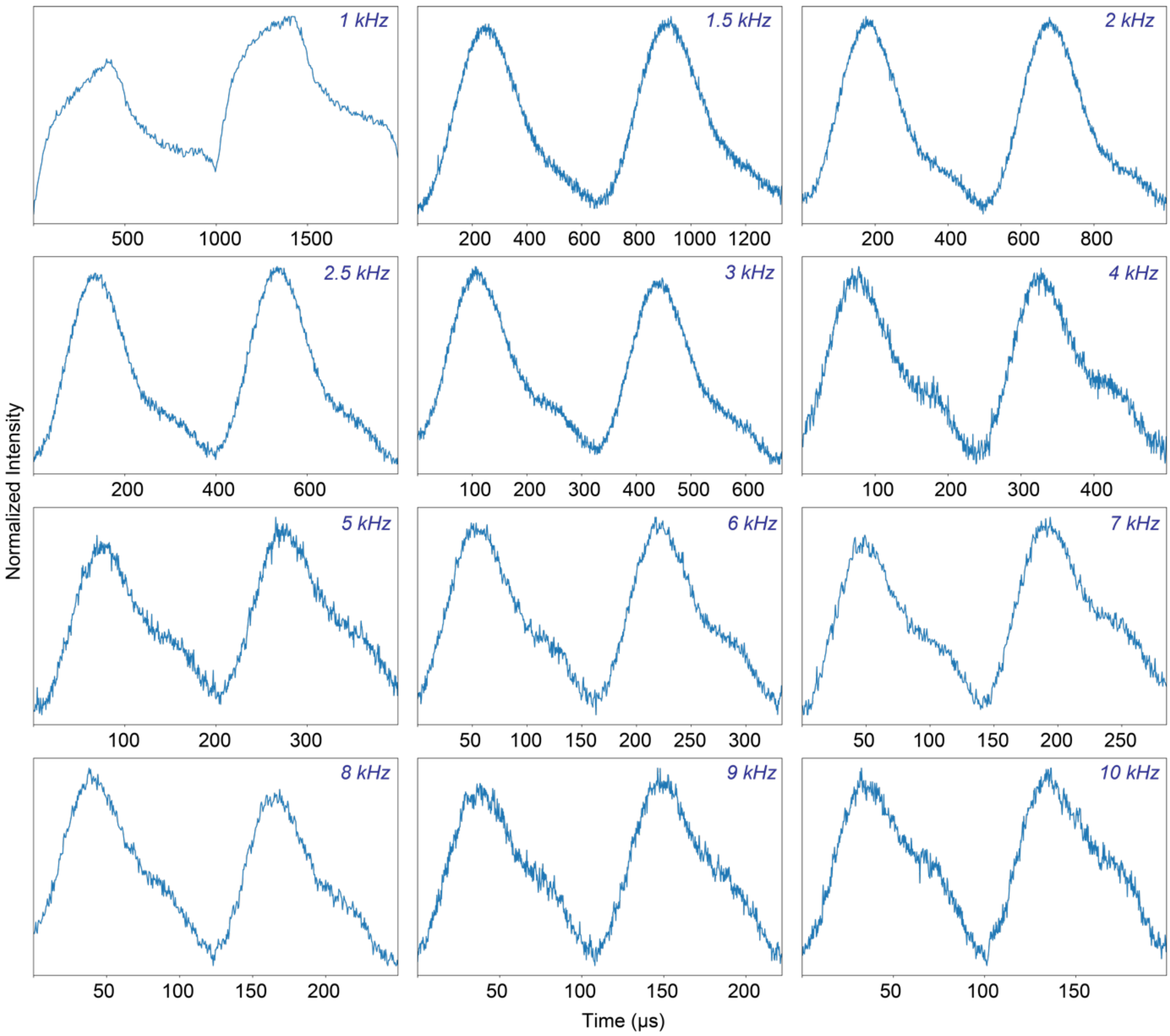

Fig. S9 The switching response of the Ga_2S_3 based optic-switch vary in control frequency at the wavelength of 532 nm.

Note S1: The calculation of average excitons lifetime

The radiative exciton lifetime can be realized in a three-phase exponential decay model.

$$y = y_0 + \sum_{i=1}^{3} (A_i \times e^{-\frac{x-x_0}{t_i}}) \quad (S1)$$

Where, the $y_0$and x_0 denote the initial values along the x and y axis, whereas the A_i represents the coefficients of three lifetime components, the t_i is the time constant for each component.

The average lifetime can be calculated using the following equation.

$$\tau_{average} = \frac{\sum(A_i \times t_i^2)}{\sum(A_i \times t_i)} \quad (S2)$$

Table S1. The radiative exciton lifetime fitting result of ultra-thin Ga_2S_3 nanoflakes

	Value
y0	0.04607
x0	27.90406
A1	54.24805
t1	0.43339
A2	0.93103
t2	2.02458
A3	0.09693
t3	9.12087
Average lifetime (ns)	**0.840**
Reduced Chi-Sqr	1.86706×10^{-5}
Adj. R-Square	0.98391

Note S2: Calculation of interference wavelength of MZI

The interference wavelength on MZI can be calculated in the following equation [1].

$$n_{eff,arm1} \cdot L_{arm1} - n_{eff,arm2} \cdot L_{arm2} = \left(m + \frac{1}{2}\right) \cdot \lambda_0 \qquad (S3)$$

Where $n_{eff,arm1}$ and L_{arm1} denote the effective refractive index and the length of the long MZI arm, respectively, whereas $n_{eff,arm2}$ and L_{arm2} represent to that of shorter arm. The m in the right side is the interference order, and the λ_0 stands for the central wavelength. In addition, the free spectral range (FSR) of the MZI device is given by [2]

$$\Delta\lambda_{FSR} = \frac{\lambda^2}{n_g \cdot \Delta L} \qquad (S4)$$

From the numerical simulation results shown in Table S2, the effective refractive index on MZI arm before and after the Ga_2S_3 deposition are 2.27433 and 2.35398, respectively. Moreover, L_{arm1} is known as 4.99 mm, while L_{arm2} is 0.30 mm. With $\lambda_0 = 1550\ nm$, the m can be calculated on a bare waveguide as follows.

$$m = \frac{2.27433 \times 4.69\ mm}{1550\ nm} - 0.5 \approx 6881 \qquad (S5)$$

Given that, the wavelength shifting before and after the deposition can be estimated as

$$\lambda_1 = \frac{2.35398 \times 4.99\ mm - 2.27433 \times 0.3\ mm}{6881 + 0.5} \approx 1607.798\ nm$$

$$\Delta\lambda_{material} = \lambda_1 - \lambda_0 \approx 57.798\ nm \qquad (S6)$$

Furthermore, as the effective refractive index on Ga_2S_3 integrated MZI arm is changed to 2.37694 under the irradiation of a control light beam, the wavelength shifting between excitation light on and off is given

$$\lambda_2 = \frac{2.37694 \times 4.99\,mm - 2.27433 \times 0.3\,mm}{6881 + 0.5} \approx 1624.447\,nm$$

$$\Delta\lambda_{light} = \lambda_2 - \lambda_1 \approx 16.649\,nm \qquad (S7)$$

Given the $\Delta\lambda_{FSR}$ calculated in Table S2, the interference wavelength will theoretically redshift by 495 periods plus ~0.083 nm after integrating material. In addition, the excitation light will further induce a redshift of 141 periods and ~0.084 nm, which is close to the measurement.

Note S3: Calculation of Ga_2S_3 coverage on MZI device

The real phase shift after the Ga_2S_3 deposition is measured as ~0.03 nm, which is about ~0.053 nm shorter than the calculation, suggesting that the long arm of MZI is not fully covered by Ga_2S_3. So, if $L_{arm1,ga2s3}$ denotes the length of the covered part on long arm, and $L_{arm1,bare}$ represents the bare part, the equation S3 can be rewritten in

$$n_{eff,ga2s3} \cdot L_{arm1,ga2s3} + n_{eff,bare}(L_{arm1,bare} - L_{arm2}) = \left(m + \frac{1}{2}\right) \cdot \lambda_{real} \qquad (S8)$$

Where, the $\lambda_{real} = \lambda_1 - 0.053 = 1607.745\ nm$, plugging the equation S8 back to the equation S3, then

$$n_{eff,ga2s3} \cdot L_{arm1,ga2s3} + n_{eff,bare} \cdot L_{arm1,ga2s3} = \left(m + \frac{1}{2}\right)(\lambda_{real} - \lambda_0)$$

$$L_{arm1,ga2s3} = \frac{(m + \frac{1}{2})(\lambda_{real} - \lambda_0)}{n_{eff,ga2s3} - n_{eff,bare}} \qquad (S9)$$

Therefore, the length of long arm with Ga_2S_3 covered is calculated as ~4.989 mm, the coverage of the Ga_2S_3 deposition can thus be estimated as ~99.98%

Table S2. The effective and group refractive indexes based on the simulation.

	Bare MZI arm	**Ga_2S_3 deposited arm**	**Ga_2S_3 deposited arm on light**
n_{eff}	2.27433	2.35398	2.37694
n_g	4.54647	4.39346	4.36024
$\Delta\lambda_{FSR}$(nm)	0.1127	0.1166	0.1175