Electronic Supplementary Material (ESI) for Nanoscale.

Supporting information

Polarized Nonlinear Optical Response in Topological Insulator Bi_2Se_3-Au Nanoantenna Hybrid-structure for All-Optical Switching

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AFM Results

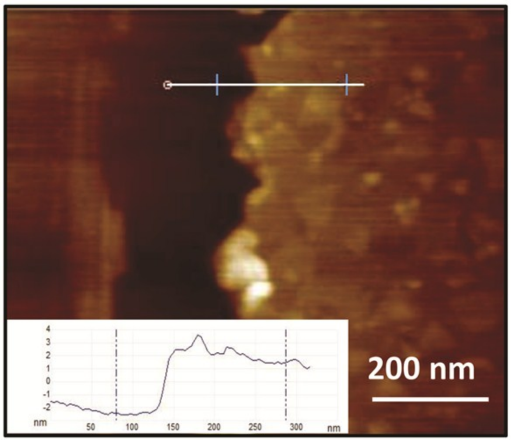

Figure S1 AFM image of the Bi_2Se_3 film shows a height of 3.895 nm which represents layers about 4.

Experimental results with ellipsometer

In the ellipsometry test, a commercial spectroscopic Mueller matrix ellipsometer (ME-L ellipsometer, Wuhan Eoptics Technology Co., Wuhan, China) is employed to investigate the optical properties of the Bi_2Se_3 film by detecting and analyzing the change in the polarization state of polarized light, whose applicable energy region covers 0.73 – 6.42 eV. The incident angle can be set among the range of 45° ~ 90°. In this experiment, the incident angles were chosen as 60°, 65°, and 70°.

In the ellipsometry analysis, a three-layer model (ambient/film/substrate) is established to fit the ellipsometry spectra. Owing to the thin thickness of Bi_2Se_3 film, the surface state is not considered. To physically embody the dielectric properties of Bi_2Se_3 over the concerned broadband range, the dielectric functions are parameterized by a combination of classical oscillators, including a five Lorentz models and two Gaussian models [1]. The measured and best matched ellipsometry spectra of Bi_2Se_3 is shown in Fig. S2(a). Obviously, the fitted ellipsometry parameters agree well with the experimental values, demonstrating the correctness of our ellipsometry analysis. Then, the real (ε_1) and imaginary (ε_2) parts of the dielectric constants are extracted and the results have been shown in Fig. S2(b). Afterwards, the complex refractive index of Real (*n*) and imaginary (*k*) parts can be calculated through the formula $N^2 = (n + ik)^2 = \varepsilon_1 + i\varepsilon_2$.

We can find that *n* racks up continuously from 300 nm to 800 nm (7.07) and then keeps a slow increment with the wavelength increasing. The imaginary parts (*k*) of index of refraction represents the ability to absorb the light. It reaches the peak about 520 nm and keep reducing above 520 nm. The strong absorption peak around 520 nm comes from the existence of the second surface state.

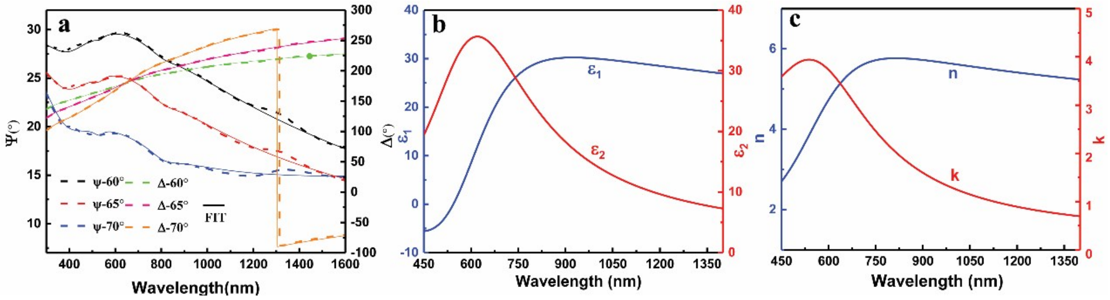

Figure. S2 a) The measured and best matched ellipsometry spectra of Bi_2Se_3, in which the dash lines and solid lines represent experiment and simulation, respectively. b) The spectral dependence of the real (ε_1) and imaginary (ε_2) parts of the dielectric constants. c) Real (*n*) and imaginary (*k*) parts of the Bi_2Se_3 index of refraction as a function of light wavelength.

Experimental results of nonlinear absorption

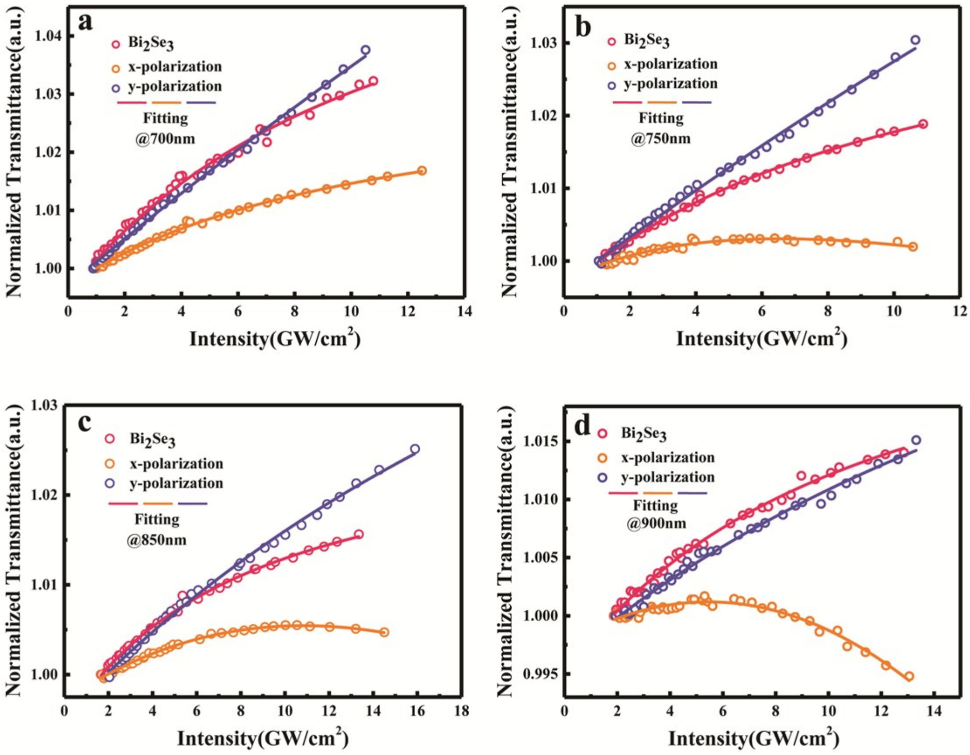

Figure S3 a), b), c), d) are the NLA data in the wavelengths of 700 nm, 750 nm, 850 nm and 900 nm respectively.

Third Nonlinear Susceptibility comparation of Au, Bi_2Se_3 and Bi_2Se_3-Au hybrid structure

According to the experimental results, the third order nonlinearity plays a dominant role in this work. Thus, the third order nonlinear susceptibility, $\chi^{(3)}$, is mathematically calculated according to the following equations [2]:

$$Re\chi^{(3)}(esu) = \frac{n_0^2 c}{12\pi^2} n_2 cm^2 W^{-1}$$

$$Im\chi^{(3)} = \left[\frac{10^{-7} c\lambda n_0^2}{96\pi^2}\right]\alpha_{NL} \tag{1}$$

Here, the refractive index n_0 is calculated by using the metamaterial homogenization technique [3]. Specifically, we calculate the homogenized refractive index for the Au-Bi_2Se_3 hybrid structure from s-parameters extracted from a series of FDTD simulations (see the equations below) [4], furnishing a frequency-dependent effective index distribution of the hybrid system.

$$s_{av} = \sqrt{s_{11}s_{22}}$$

$$z = \sqrt{\frac{(1 + s_{av}^2 - s_{21}^2)}{(1 - s_{av})^2 - s_{21}^2}}$$

$$neff = \frac{-i}{2\pi d/\lambda}\log\left[\frac{s_{21}}{1 - s_{av}\frac{z-1}{z+1}}\right]$$

(2)

On the other hand, α_{NL} is extracted through the nonlinear absorption experiments with the formula $dI/dL = -(\alpha_0 + \alpha_{NL}I)I$ [2]. Another important factor, the nonlinear refractive index n_2, is acquired by dividing the time-averaged refractive index by the incident intensity [4].

Take the resonance of 800nm for instance, the relevant nonlinear parameters are listed as follows:

Supplementary Table 1. Comparison of third nonlinear susceptibility of Au, Bi_2Se_3 and Bi_2Se_3-Au hybrid structure.

Sample	n_0	n_2**(cm²/W)**	α_{NL} **(cm/GW)**	$Re\chi^{(3)}$**(m²/V²)**	$Im\chi^{(3)}$**(m²/V²)**
Bi_2Se_3-Au (x-polarization)	6.0975	-2.84×10^{-10}	-549	-4.2021×10^{-16}	-8.1310×10^{-18}
Bi_2Se_3-Au (y-polarization)	5.5619	-9.15×10^{-11}	-6606.7	-1.1265×10^{-16}	-8.1414×10^{-17}
Bi_2Se_3	5.7658	-2.26×10^{-10}	-3797.2	-2.9234×10^{-16}[5]	-5.0340×10^{-17}
Gold	--	--	--	1.1339×10^{-16}[6]	4.7284×10^{-17}[6]

Ultra-fast carrier's relaxation process

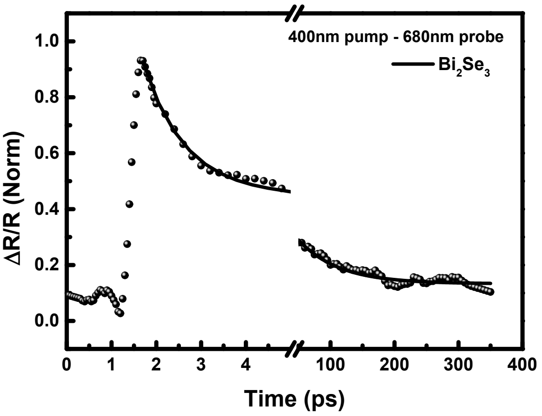

Figure S4 The normalized transient reflection change of 4-QL Bi_2Se_3 film in the long-time range of 0-350 ps, showing the carriers behavior of the sample.

Reference

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