Supporting information

Polarized Nonlinear Optical Response in Topological Insulator

Bi$_2$Se$_3$-Au Nanoantenna Hybrid-structure for All-Optical Switching

Runlin Miao,$^a$ Yuze Hu,$^a$ Hao Ouyang,$^a$ Yuxiang Tang,$^a$ Chenxi Zhang,$^a$ Jie You,$^b$ Xin Zheng,$^b$
Zhongjie Xu,$^a$ Xiang’ai Cheng$^a$ and Tian Jiang$^*$

$^a$College of Advanced Interdisciplinary Studies, National University of Defense Technology,
    Changsha 410073, China
$^b$National Innovation Institute of Defense Technology, Academy of Military Sciences PLA China,
    Beijing 100071, China
†These authors contributed equally to this work.
*Corresponding author: tjiang@nudt.edu.cn.

AFM Results

Figure S1 AFM image of the Bi$_2$Se$_3$ 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$_2$Se$_3$ 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$_2$Se$_3$ film, the surface state is not considered. To physically embody the dielectric properties of Bi$_2$Se$_3$ 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$_2$Se$_3$ 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 (\(\varepsilon_1\)) and imaginary (\(\varepsilon_2\)) 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$_2$Se$_3$, in which the dash lines and solid lines represent experiment and simulation, respectively. b) The spectral dependence of the real (\(\varepsilon_1\)) and imaginary (\(\varepsilon_2\)) parts of the dielectric constants. c) Real (\(n\)) and imaginary (\(k\)) parts of the Bi$_2$Se$_3$ 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 comparison of Au, Bi$_2$Se$_3$ and Bi$_2$Se$_3$-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]:

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

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

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$_2$Se$_3$ 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.

$$s_{uv} = \sqrt{s_{11}s_{22}}$$
\[
\begin{align*}
\frac{\gamma}{(1+\alpha_{av})(1-\alpha_{av})} &= \frac{1}{N_{eff}^2} \\
&= -i \frac{\alpha_{av}}{2\pi d/\lambda} \log \left( \frac{\gamma_2}{1-\frac{\alpha_{av}}{\gamma}+1} \right) \\
\end{align*}
\]

(2)

On the other hand, \(\alpha_{NL}\) is extracted through the nonlinear absorption experiments with the formula \(dI/dt = -(\alpha_0 + \alpha_{NL})I\) [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\(_2\)Se\(_3\) and Bi\(_2\)Se\(_3\)-Au hybrid structure.

<table>
<thead>
<tr>
<th>Sample</th>
<th>(n_0)</th>
<th>(n_2(\text{cm}^2/\text{W}))</th>
<th>(\alpha_{NL}) (cm/GW)</th>
<th>(Re\chi^{(3)}(\text{m}^2/\text{V}^2))</th>
<th>(Im\chi^{(3)}(\text{m}^2/\text{V}^2))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bi(_2)Se(_3)-Au (x-polarization)</td>
<td>6.0975</td>
<td>(-2.84 \times 10^{-10})</td>
<td>-549</td>
<td>(-4.2021 \times 10^{-16})</td>
<td>(-8.1310 \times 10^{-18})</td>
</tr>
<tr>
<td>Bi(_2)Se(_3)-Au (y-polarization)</td>
<td>5.5619</td>
<td>(-9.15 \times 10^{-11})</td>
<td>-6606.7</td>
<td>(-1.1265 \times 10^{-16})</td>
<td>(-8.1414 \times 10^{-17})</td>
</tr>
<tr>
<td>Bi(_2)Se(_3)</td>
<td>5.7658</td>
<td>(-2.26 \times 10^{-10})</td>
<td>-3797.2</td>
<td>(-2.9234 \times 10^{-16})</td>
<td>([-5.0340 \times 10^{-17}]</td>
</tr>
<tr>
<td>Gold</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>1.1339 \times 10^{-16}[6]</td>
<td>4.7284 \times 10^{-17}[6]</td>
</tr>
</tbody>
</table>

**Ultra-fast carrier’s relaxation process**

![Figure S4](image)

**Figure S4** The normalized transient reflection change of 4-QL Bi\(_2\)Se\(_3\) film in the
long-time range of 0-350 ps, showing the carriers behavior of the sample.

Reference


