Electronic supplementary information (ESI)

Role of Carrier-Transfer in the Optical Nonlinearity of Graphene/Bi_2Te_3 Heterojunctions

Jia-Chi Lan,†[a] Junpeng Qiao,†[a] Wei-Heng Sung,†[a] Chun-Hu Chen,*[b] Ren-Huai Jhang,[b] Shi-Hsin Lin,[c] Li-Ren Ng,[c] Gengchiau Liang,[d] Meng-Yu Wu,[a] Li-Wei Tu,[e] Cheng-Maw Cheng,[f] Hong Liu, *[g] and Chao-Kuei Lee*[aeh]

[a]Department of Photonics, National Sun Yat-sen University, Kaohsiung, 80424, Taiwan.

E-mail: chuckcklee@yahoo.com

[b]Department of Chemistry, National Sun Yat-sen University, Kaohsiung, 80424, Taiwan.

E-mail: chunhu.chen@mail.nsysu.edu.tw

[c]Department of Materials and Optoelectronic Science, National Sun Yat-sen University, Kaohsiung, 80424, Taiwan.

[d]Department of Electrical and Computer Engineering, National University of Singapore, Singapore, 117583, Singapore.

[e]Department of Physics and Center of Crystal Research, National Sun Yat-sen University, Kaohsiung, 80424, Taiwan

[f]National Synchrotron Radiation Research Center, Hsinchu, 30076, Taiwan.

[g]State Key Laboratory of Crystal Materials, Shandong University, Jinan, 250100, China

E-mail: hongliu@sdu.edu.cn

[h]Research Center for Applied Sciences, Academia Sinica, Taipei, 11529, Taiwan

†These authors contributed equally.

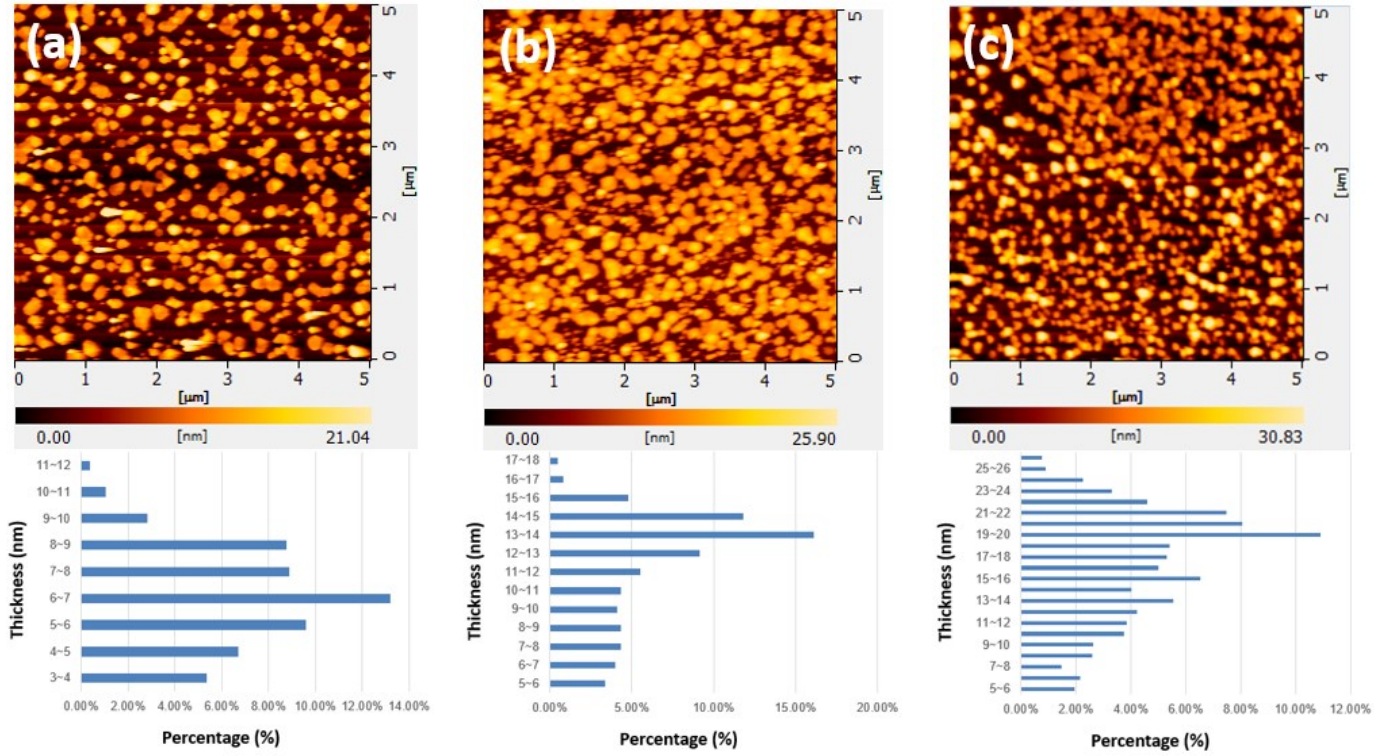

Fig. S1. AFM image and thickness distribution of (a) 6.5 nm, (b) 12.3 nm and (c) 16.5 nm Bi_2Te_3 nanoplates.

X-ray photoelectron spectrum (XPS) experiment was conducted by JEOL JAMP-9500F using Mg Kα X-ray. Fig. S2 (a) is the wide range XPS of graphene/Bi_2Te_3 heterojunction. Fig. S2 (b), (c)

and (d) are the deconvolution of C 1s, Bi 4f and Te 3d peak. In Fig. S2 (b), a peak is located at 284.03 eV, indicating the graphene C-C bonding. The Bi $4f_{7/2}$ spectrum is shown in Fig. S2 (c), there is one peak located at 158.73 eV, indicating the Bi_2Te_3 Bi-Te bonding. In Fig. S2 (d), beside the main peaks which located at 575.63 eV, the small components at 571.69 eV is stemming from surface oxidation which is commonly observed in synthesized topological insulator. Though there is little amount of oxidized state in Bi_2Te_3 nanoplate, they would not dramatically affect the Bi_2Te_3 electronic property. Similar results have been reported in previous study.[1] Therefore, one can conclude that a high-quality graphene/Bi_2Te_3 heterojunction with little oxidation was successfully fabricated.

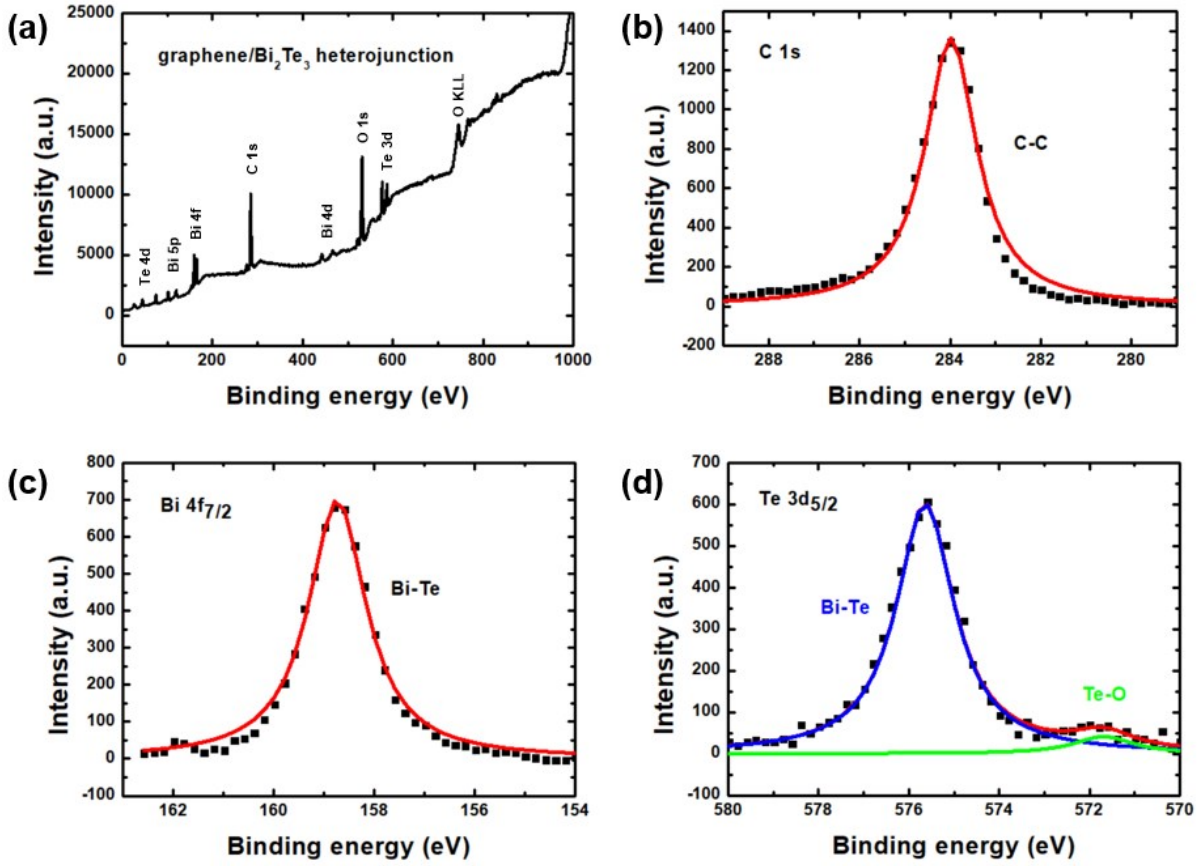

Fig. S2. (a) XPS for heterojunction of monolayer graphene with Bi_2Te_3. (b) The C 1s peak, (c) the Bi 4f peak and (d) the Te 3d peak.

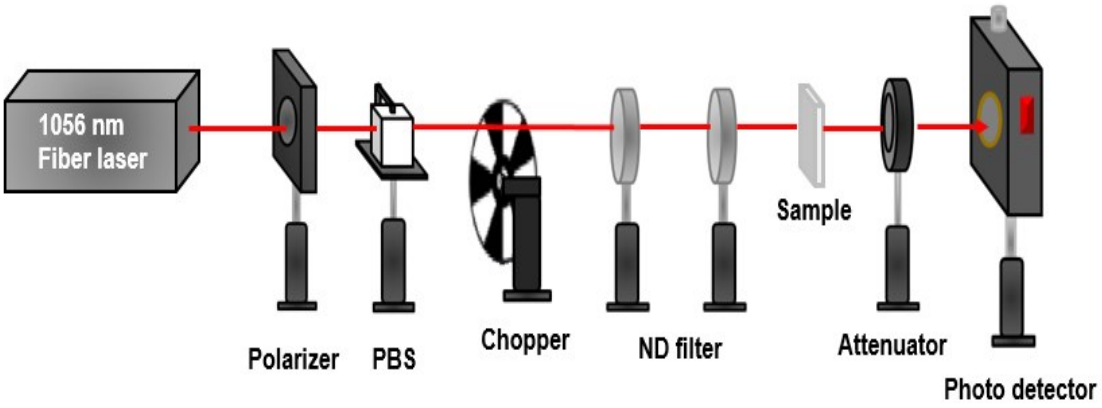

Fig. S3. Schematic of saturable absorption measurement.

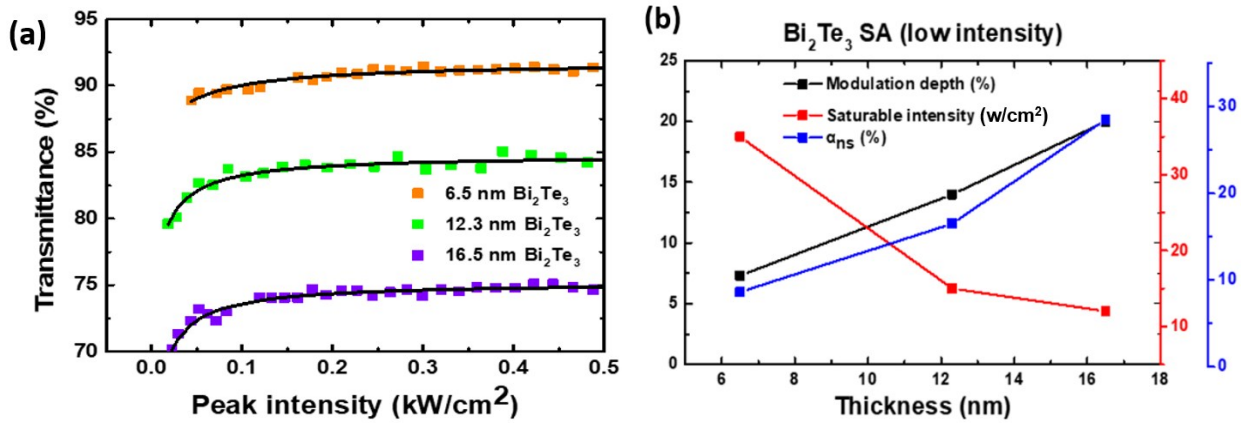

Fig. S4. (a) Nonlinear transmittances and fitting curves of 6.5 nm, 12.3 nm and 16.5 nm Bi_2Te_3 heterojunction at low optical intensity. (b) Fitting values of modulation depth, saturable intensity and non-saturable absorption coefficient at low intensity versus Bi_2Te_3 nanoplate thickness.

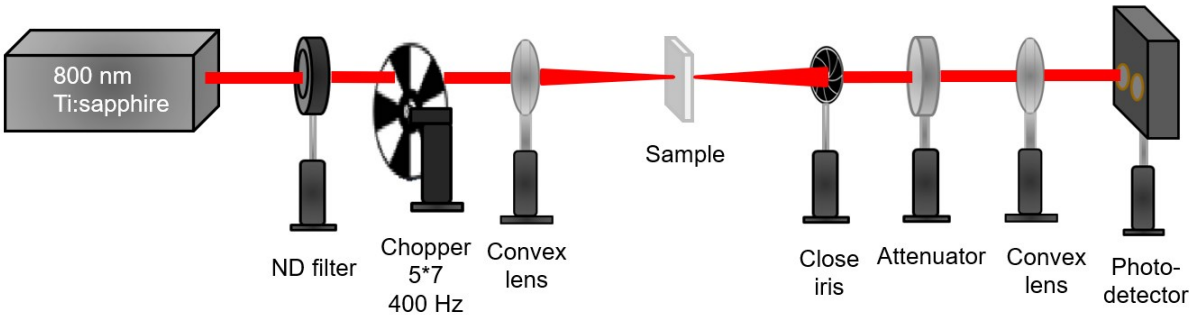

Fig. S5. Schematic of z-scan measurement.

The close aperture/open aperture data is fitted with conventional equation as bellow:

$$T_{CA/OA} = 1 + \frac{4 \times \frac{Z}{Z_R} \times \Delta\emptyset_0}{((\frac{Z}{Z_R})^2 + 1)((\frac{Z}{Z_R})^2 + 9)}$$

Where the Z is the sample position, $^{\Delta\emptyset_0}$ is nonlinear phase shift, $Z_R = \pi\omega_0^2/\lambda$ is Rayleigh length, ω_0 is beam radius at focal point about 86.3 μm, λ is laser wavelength. The $^{\Delta\emptyset_0}$ of graphene/6.5 nm Bi_2Te_3 heterojunction under 98 MW/cm^2 is -0.028. Finally, the nonlinear refractive index n_2 is:

$$n_2 = \frac{\Delta\emptyset_0}{I_0 \times k \times L}$$

Where the I_0 is peak intensity at focal point, k is wavenumber, L is sample thickness. The n_2 is then calculated about -5.28×10^{-9} cm^2/W.

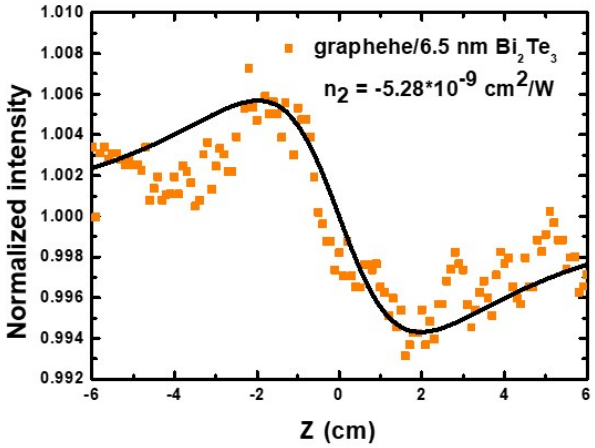

Fig. S6. Close aperture/open aperture z-scan result of graphene/6.5 nm Bi_2Te_3 under 98 MW/cm^2.

Computation of graphene/Bi_2Te_3 heterojunction binding energy

Our first-principles calculations was performed via Vienna Ab-initio Simulation Package (VASP) to obtain the binding energy.[2,3] Perdew-Burke-Ernzerhof (PBE) functional and the van der Waals D3 correction were employed to describe the exchange-correlation energy.[4,5] We also considered the spin-orbit coupling in our calculation as Bi and Te atoms are heavy elements. In our calculation, we computed the adsorption energy within the heterojunction Bi_2Te_3 - graphene in mono-quintuple layer (QL) and two QLs of Bi_2Te_3. A 2×2×1 supercell of graphene with 8 carbon atoms was stacked above the QL of Bi_2Te_3, resulting 10.88% strain for Bi_2Te_3. The optimized crystal structure is shown in Figure S4. The binding energy of the heterojunction graphene/mono-QL Bi_2Te_3 is -0.292 eV and the graphene/two-QLs Bi_2Te_3 is -0.278 eV.

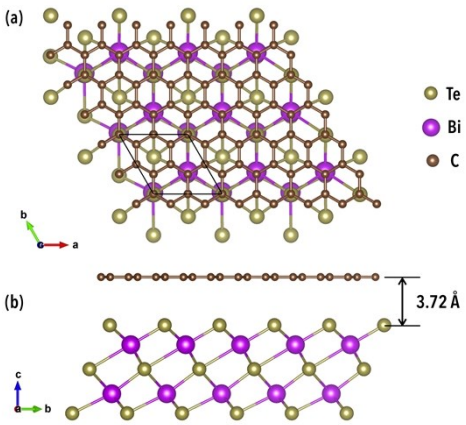

Fig. S7 (a) Top view and (b) side view of the equilibrium structure of mono-QL Bi_2Te_3 stacking with graphene.

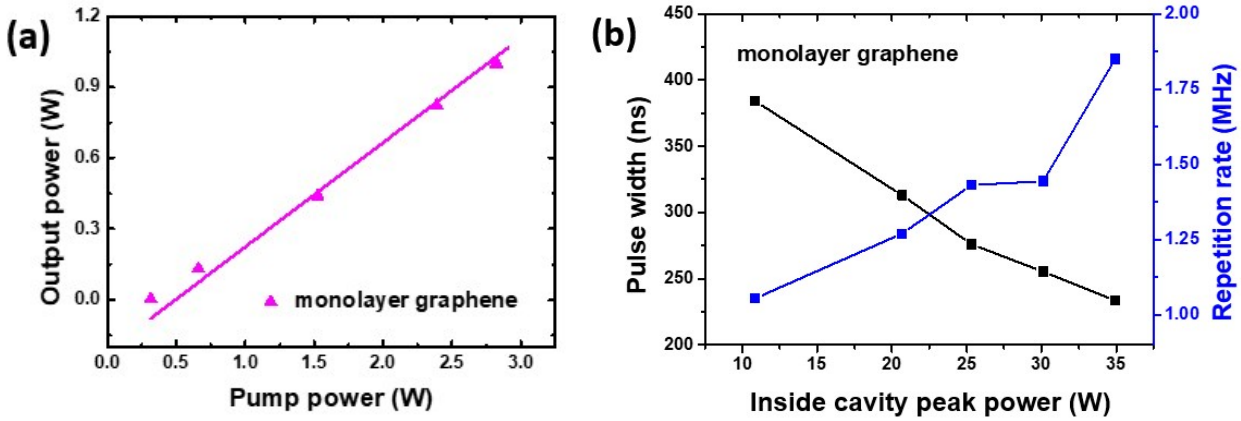

Fig. S8 (a) Average output power versus pump power of graphene. (b) Pulse width and repetition rate versus inside cavity peak power of graphene.

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