Supporting Information

Title: 2D Graphdiyne: excellent ultraviolet nonlinear absorption material

Fang Zhang[1§], Guowei Liu[1], Junjie Yuan[1], Zhengping Wang[2], Tianhong Tang[2], Shenggui Fu[1], Huanian Zhang[1], Zhongsheng Man[1], Fei Xing[1§] and Xinguang Xu[2]

§ Corresponding author: yourszhangfang1988@126.com, xingfei@sdut.edu.cn.

Table of contents:

S1. Preparation processes of graphdiyne nanosheets.

Adopting liquid phase exfoliation (LPE) method, graphdiyne nanosheets were prepared. The preparing processes is exhibited in figure S1. Firstly, the graphdiyne powders were grinded for 5 hours and then dissolved in ultra-pure alcohol. A ultrasonic treatment with 40 KHz frequency was carried out for dispersing the nanosheets of the dispersion liquid. Then the dispersion liquid was centrifugalized for 30 minutes, which made the dispersion liquid stratified. One fifth of the upper dispersion liquid was collected. One part of the dispersion liquid was stored for the nonlinear absorption measurement. Another part was used for the sample characterization. The sample concentration was estimated to be about 5 mg/L.

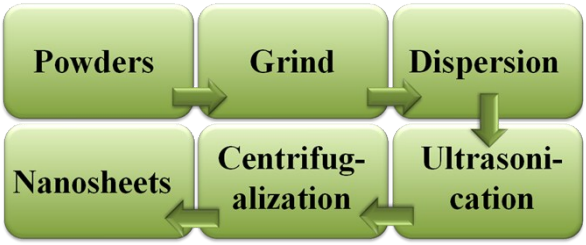

Figure S1. LPE method for 2D nanosheets.

S2: Energy-dispersive X-ray spectroscopy (EDS) result of GDY powder and Raman spectrum of graphene.

In order to compare the Raman properties of graphdiyne powder, the Raman spectrum of graphene powder was carried out and shown in fig. S2. The wave-numbers of D and G bands are 1344 and 1580 cm^{-1}, which are associated with the vibrations of C atoms with sp^3 electronic configuration and in-plane vibrations of sp^2-bonded C atoms, respectively.

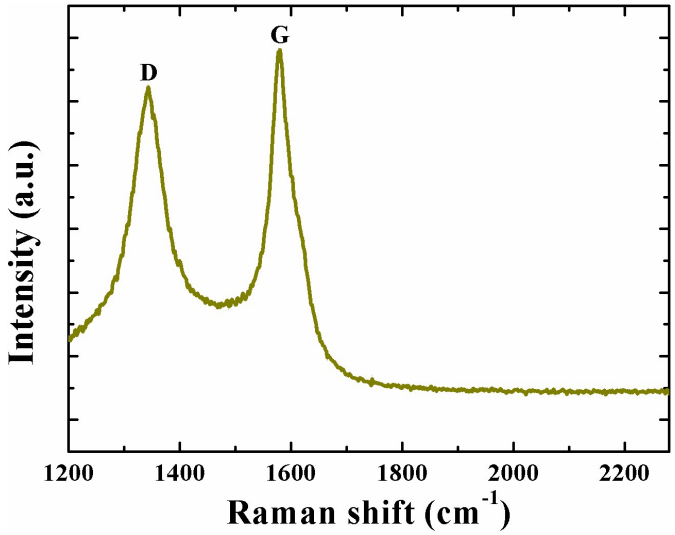

Figure S2. Raman spectrum of graphene.

S3: Z-scan setup.

The nonlinear absorption (NLA) experiment was detected using open-aperture Z-scan method with nanosecond and picosecond exciting lasers, respectively. The experimental setup is shown in figure S3.

(a) The ns Z-scan setup

The excited laser is an OPO laser with tuning range of 192~2750 nm (Continuum Inc, America). The pulse width and the repetition rate are 6 ns and 10 Hz, respectively. Four lasers with wavelengths at 290, 410, 532 and 1064 nm were selected as light sources. The focal length of the convex lens is 30 mm at 1064 nm. The prepared dispersion liquid was shored in a quartz colorimetric utensil with 2 mm thickness and was placed in **Z**-position, which could move in z-direction.

(b) The ps Z-scan setup

Different from the ns Z-scan setup, the laser resource of ps Z-scan setup is a dye mode-locked Nd:YAG laser (PY61C-10, Continuum Inc, America). The wavelength is 1064 nm, the pulse width is 40 ps and the repetition rate is 10 Hz. In order to obtain other excited lasers, the frequency conversion technology was adopted. A KTP crystal was selected for the generation of 532 nm laser (frequency doubling laser). Mirror M_1 is a filter with high transmissivity at 532 nm (T>80%) and high reflectivity at 1064 nm (R>99.5%). The pulse width of 532 nm wavelength is around 30 ps. In addition, using the sum-frequency technology of 1064 and 532 nm lasers, the third harmonic laser with 355 nm wavelength was generated with BBO crystal. Mirror M_2 is the filter with high transmissivity at 355 nm (T>60%) and high reflectivity at 1064, 532 nm

(R>99.5%). The pulse width of 355 nm wavelength is around 20 ps.

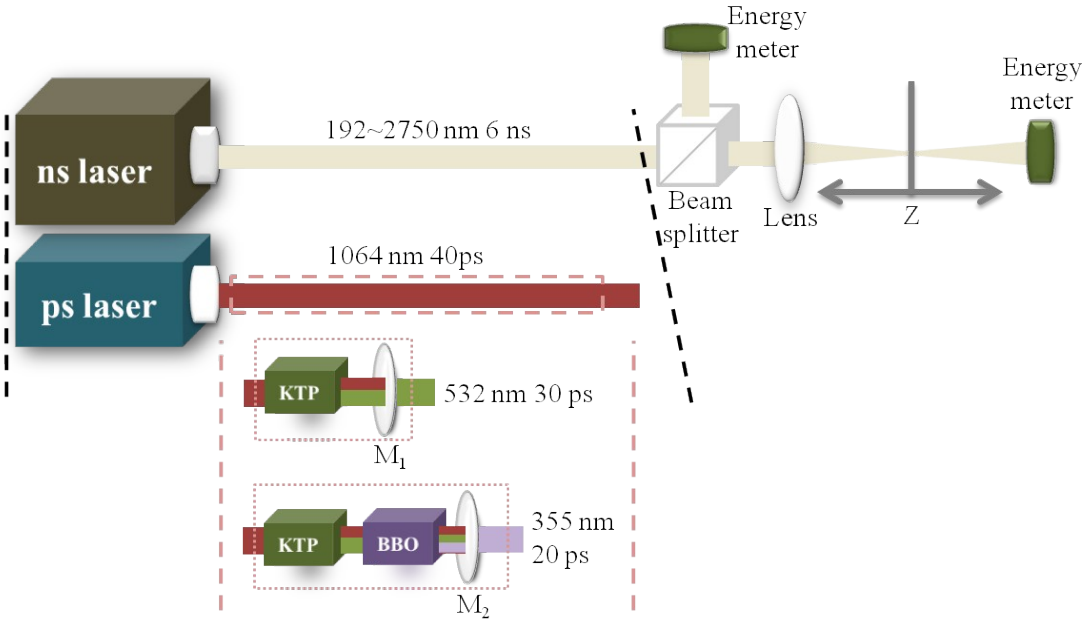

Figure S3. Open-aperture Z-scan experimental setup with ns and ps setup.

S4: NLA results with 1300, 1800 and 2100 nm wavelength excitation in ns setup.

When the excited wavelengths are 1300, 1800 and 2100 nm, the NLA results are shown in Fig. S4 with intensity of 280, 230 and 100 μJ, respectively. The normalized transmittances are maintain as a constant 1 and no NLA behaviors can be observed at these excited wavelengths and intensities. According to our results with 290, 410, 532 and 1064 nm excitation, in infrared waveband, the NLA behavior calls for much higher excited intensity. At the same time, the output intensity of OPO laser in infrared waveband becomes lower. So no apparent NLA behavior could be observed with the excitation of 1300, 1800 and 2100 nm wavelengths due to the such high excited intensity and reachless output intensity of laser.

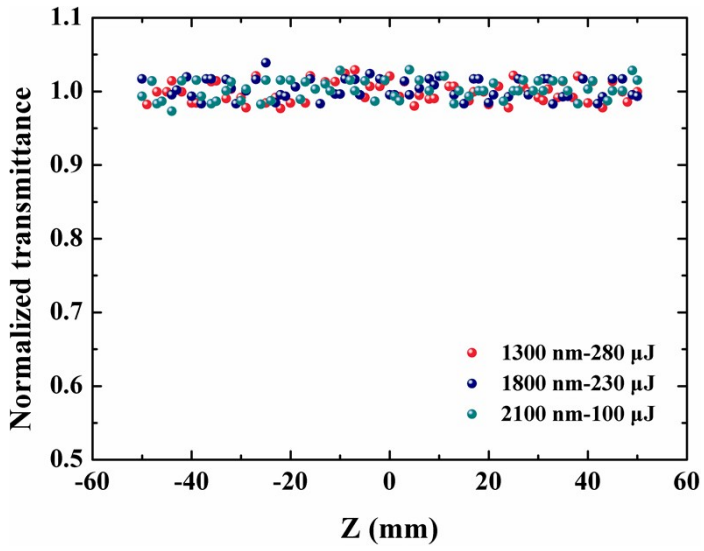

Figure S4. NLA results with 1300, 1800 and 2100 nm wavelengths excitation.