**Supporting information**

**The pump fluence and wavelength dependent ultrafast carrier dynamics and optical nonlinear absorption in black phosphorus nanosheets**

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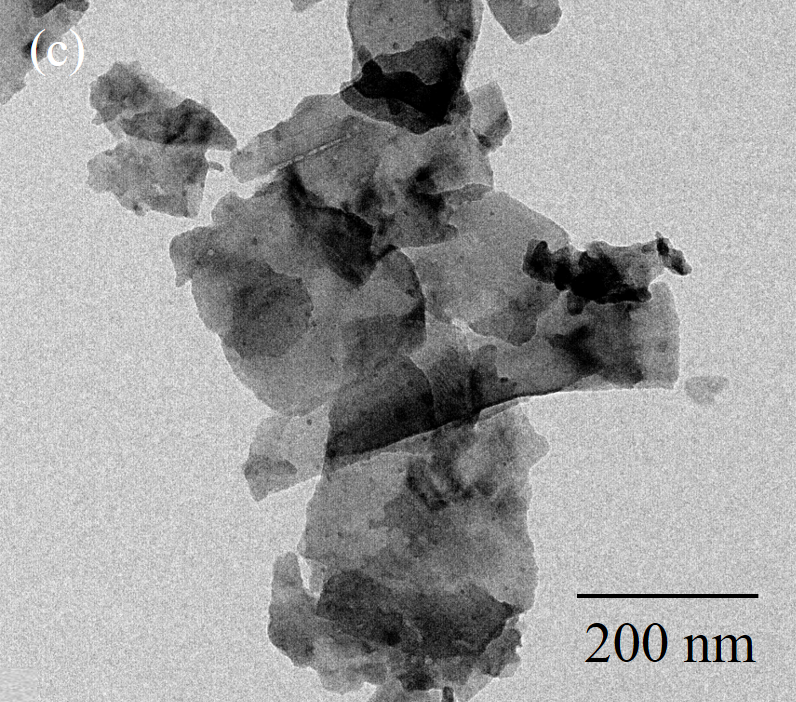
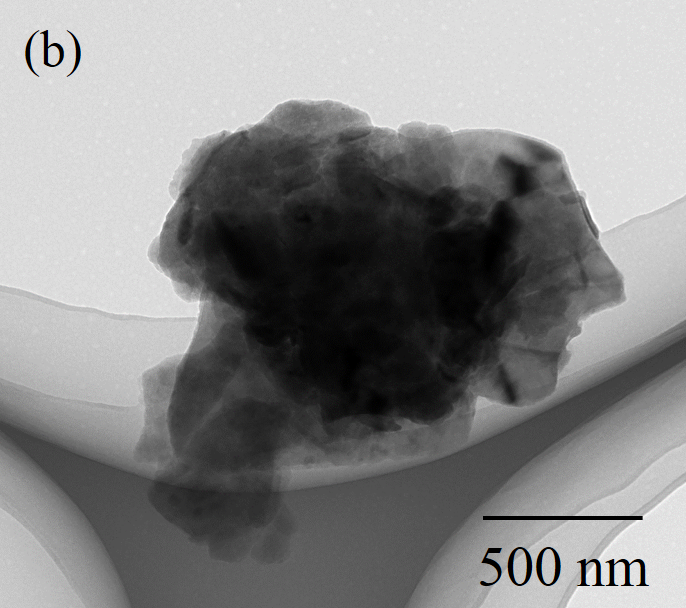
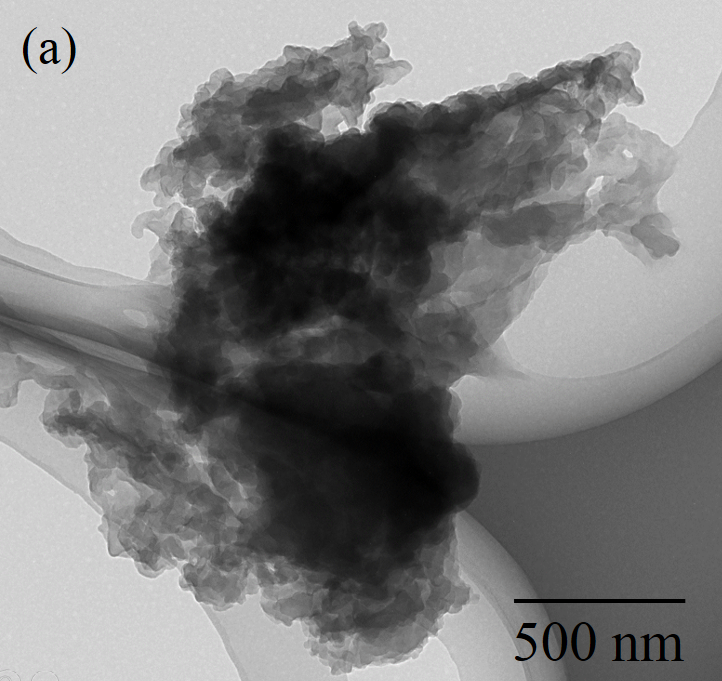
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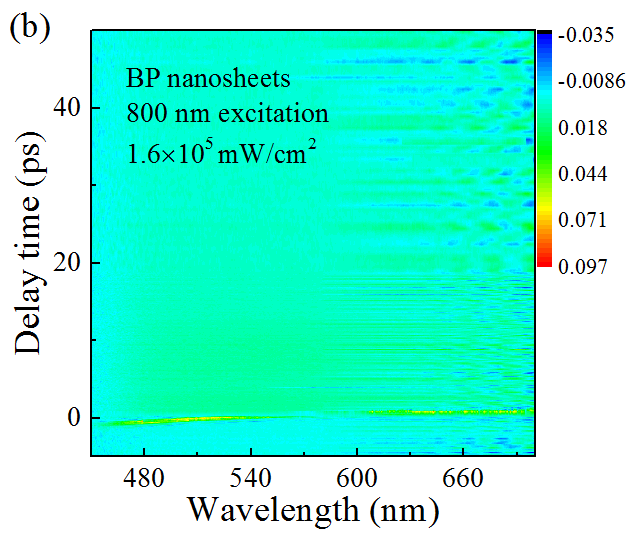
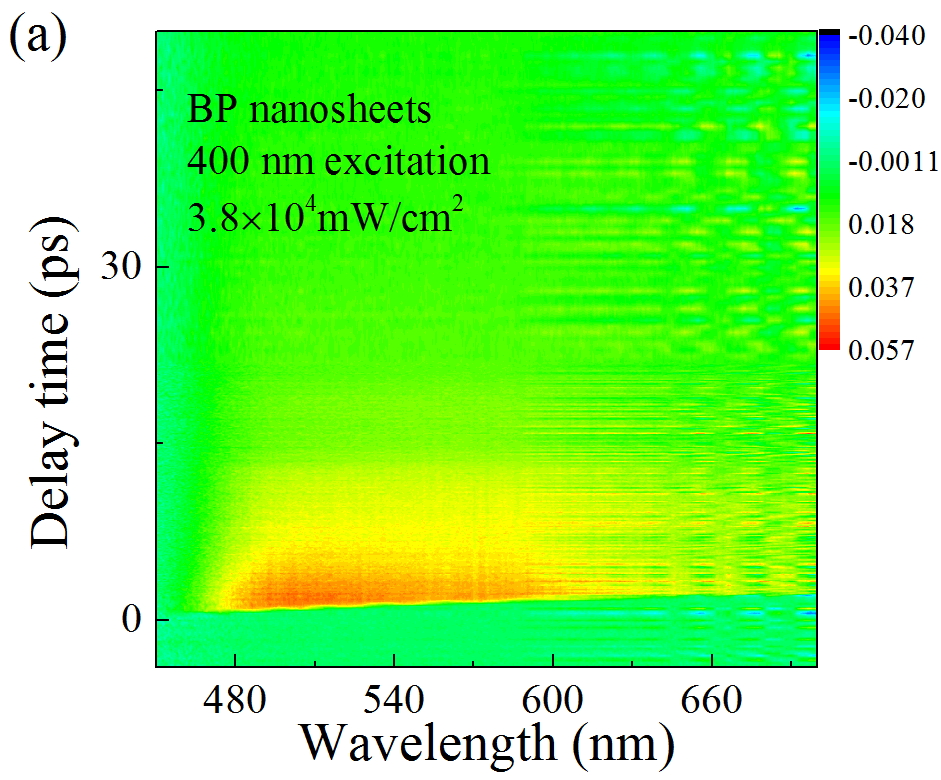
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**Fig. S1** (a) The absorbance spectrum. (b) The corresponding Tauc plot.The quasi-bandgap (*Eg*) can be estimated to be about 1.90~2.0 eV.



**Fig. S2** TEM images of BP nanosheets at different locations and scale bars



**Fig. S3** (a)The 2D mapping of TA spectra for BP nanosheets pumped at 400 nm with fluence of 3.8×104 mW/cm2. (b) The 2D mapping of TA spectra for BP nanosheets pumped at 800 nm with fluence of 1.6×105 mW/cm2.



**Fig. S4** Nanosecond open-aperture Z scan measurement for (a) carbon disulfide at 520 nm under the peak fluence of 15.6 GW/cm2 (blue square) and 20.4 GW/cm2 (red square), respectively, and (b) water (used as solvent for BP nanosheets) at 520 nm under the peak fluence of 10.5 GW/cm2.

**Tab. S1** Absorption coefficient index with the pump fluence in Fig. 4

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| *λ* | Pump fluence | *T* | α | *Is* | *β* |
| nm | GW/cm2 | % | cm-1 | (×10-2) GW/cm2 | cm/GW |
| 480 | 0.22 | 36.4 | 5.053 | 7.2 | - |
| 520 | 0.11 | 35.1 | 5.235 | 5.7 | - |
| 0.36 | 35.1 | 5.235 | 7.3 | - |
| 0.64 | 35.1 | 5.235 | 8.8 | 4.9 |
| 530 | 0.22 | 33.0 | 5.543 | 7.1 | - |
| 580 | 0.22 | 65.7 | 2.100 | 10.5 | - |
| 640 | 0.22 | 70.0 | 1.783 | 24.1 | - |
| 680 | 0.22 | 69.4 | 1.826 | 28.4 | - |

**Abnormal carrier lifetime measured at 520 nm for ultrafast dynamics under 800 nm**

The ultrafast dynamics at 800 nm can be fitted with the single exponential decay model as,

 (1)

where *τ* is the lifetime, *τfwhm* is the instrumental response function, A represents the baseline of signal and B represents the amplitude.

The probe wavelength dependent lifetime with pump fluence at 8.4×104 mW/cm2 is shown in Fig. S5 below. The error bar here represents the standard deviation from the fitting process. With longer wavelength, the lifetime shows an increasing trend. However, the fitted lifetime at 520 nm is larger than the one at 540 nm. The abnormal carrier lifetime at this particular wavelength can be interpreted in two ways:

1. The error from the experiment (temperature, humidity, laser stability) at these two wavelengths is larger than 1 ps. Since the error bar presented only accounts for the error from the fitting process, the true error (experiment error + fitting error) could be larger. So, it is still possible that the true lifetime at 540 nm is larger than the one at 520 nm. To verify this, multiple experiments have to be carried out at different days with different experimental conditions. However, the status of solution might also change if the measurement needs really long time (if the period of the error from experiment is very long). So, it is better to conduct the experiment by a laser with much more stable condition to see if the fitted time would change. Considering the fluctuations caused by all factors, the overall trend of lifetime still increases with probe wavelength.

2. The lifetime at 520 nm is indeed larger than the one at 540 nm. From Figure 4(C) and (D), there are two processes happening. For different wavelengths at the same pump fluence, the lifetime shows increasing trend at larger wavelength. For the same wavelength and different pump fluences, the lifetime first increases and then decreases. At extremely large pump fluence, the lifetime should increase asymptotically approaching the one at 400 nm (from the hypothesis in the main text). At 520 nm with pump fluence at 8.4×104 mW/cm2, the lifetime increases compared with that at low fluence because of the two-photon absorption. Compared with the lifetime measured at 540 nm, the slightly longer lifetime at 520 nm (at the fluence of 8.4×104 mW/cm2) might be the result of the competition between two-photon absorption and saturation effect as discussed in the main text. In another word, since the lifetime at one wavelength first increases and then decreases with increasing the fluence, for 520 nm, the effective lifetime might be just around the peak position with the fluence at 8.4×104 mW/cm2. To prove this, further experimental investigation is necessary to scan the region from 500 to 540 nm with much smaller spectrum width. Also, the wavelength dependent lifetime needs to be measured under different fluences.



**Fig. S5** Probe wavelength dependent lifetime of BP nanosheets (pump at 800 nm) with pump fluence at 8.4×104 mW/cm2.