Nonlinear absorption in two dimensional materials

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Abstract
Thin film materials with large optical nonlinearity and ultrafast response are required as the promising candidates for future integrated photonics applications. Recently, the two dimensional layered transition metal dichalcogenides (TMDs) and black phosphorus (BP) have attracted much attention due to their unique electronic, optical, mechanical, chemical and thermal properties. WS_2 and MoS_2 mono- and few-layer films were fabricated by vapor phase sulfurization and chemical vapor deposition method. We investigated the optical nonlinearity of mono- and few-layer WS_2 and MoS_2 using the Z-scan technique at a wavelength of 1030 nm, 800 nm and 515 nm with femtosecond pulses. The monolayer WS_2 exhibited giant optical nonlinearities having the two-photon absorption (TPA) coefficient of ~1.0×10^4 cm/GW. The layer number and excitation wavelength dependence of the optical nonlinearity of WS_2 and MoS_2 were investigated. The mechanism of the optical nonlinearity was discussed in detail. The damage thresholds of TMDs were also given to support potential device application in the future. We have obtained the ground-state absorption (GSA) and excited-state absorption (ESA) cross section σ_{gs} and σ_{es} of BP nanosheets, 1.25×10^{-16} cm^2 and 2.85×10^{-17} cm^2 at 515 nm, 1.23×10^{-17} cm2 and 3.02×10^{-18} cm^2 at 1030 nm with the slow saturable absorber (SA) model. The ratio of σ_{es}/σ_{gs} is about 0.228 at 515 nm and 0.247 at 1030 nm. It is even lower when the fast SA model is applied. The σ_{gs} and σ_{es} of MoS_2 and graphene were also achieved using the slow SA model. It is clear that the ratio σ_{es}/σ_{gs} of BP is the smallest among three two-dimensional layered materials. The superior saturable absorption performance of BP over MoS2 and graphene can be well explained from this perspective. The results fundamentally support BP to be used as a saturable absorber in nanophotonic devices, such as fiber laser mode-locking, Q-switching, optical switches, etc.

References
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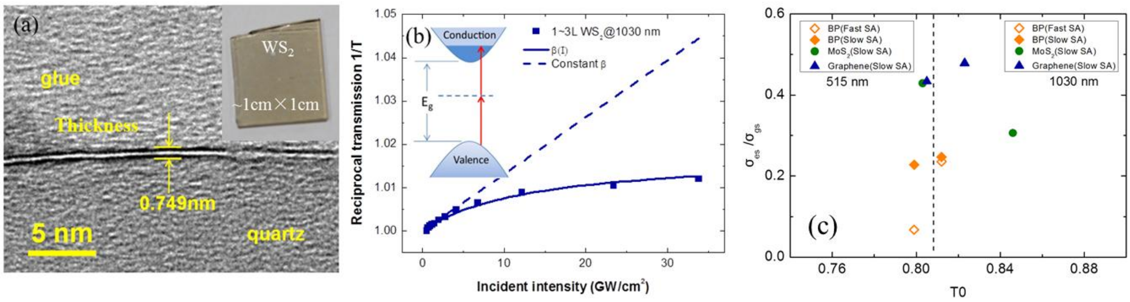

Fig. 1 (a) Cross-sectional TEM of the 1~3L WS_2 film, (b) Reciprocal transmission versus irradiance of 1~3L WS_2 film. Solid squares are the measured data. Solid line: theoretical variation for a hyperbolic irradiance dependence of the TPA coefficient. Dashed line: theoretical variation for a constant TPA coefficient. (c) The ratio of ground-state absorption and excited-state absorption cross section of BP, MoS2 and graphene at 515 nm and 1030 nm.