Ultrafast Nonlinear Optical Effects in 2D Semiconductors

Jun Wang
Key Laboratory of Materials for High-Power Laser, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai 201800, China
jwang@siom.ac.cn

As a class of semiconductors, transition metal dichalcogenides (TMDs) have the formula MX_2, where M stands for a transition metal (i.e., Mo, W, Ti, Nb, etc.) and X stands for a chalcogen (i.e., S, Se or Te). TMDs show graphene-like layered structure. Strong covalent bonds in layers and weak van der Waals interaction between layers allow TMDs to form a robust 2D nanostructure. In a TMD monolayer, the single transition metal layer is sandwiched between the two chalcogen layers. Owing to the specific 2D confinement of electron motion and the absence of interlayer coupling perturbation, 2D layered TMDs show unique photonics-related physical properties, e.g.,

1) Sizable and layer-dependent bandgap, typically in the 1-2 eV range;
2) Indirect-to-direct bandgap transition as the decreasing of the number of monolayer;
3) Fairly good photoluminescence and electroluminescence properties;
4) Remarkable excitonic effects, i.e., high binding energy, large oscillator strength and long lifetime.

In combination of the ultrafast carrier dynamics and molecular-scale thickness, the prominent properties manifest the 2D TMDs a huge potential in the development of photonic devices and components with high performance and unique functions.

We have extensively studied the ultrafast nonlinear absorption and nonlinear refraction of layered MX_2 (X=S, Se, Te) over broad wavelength (Vis-NIR) and time (fs-ps-ns) ranges [1-9]. Large area MoS_2 neat films with controllable thicknesses were fabricated from liquid-exfoliated MoS_2 dispersions by vacuum filtration. The MoS_2 films show superior broadband ultrafast saturable absorption (SA) performance, in comparison with the graphene films and the MoS_2 dispersions [4]. Very recently, we observed giant two-photon absorption (TPA) coefficient in a WS_2 monolayer [5]. The order of magnitude of TPA coefficient in WS_2 monolayer (~100 cm/MW) exceeds that of the conventional semiconductors (e.g., CdTe, GaAs, ZnS, ZnO, etc.) by a factor of 3-4. This is also the first Z-scan performance on an optical medium with a thickness as tiny as 0.75 nm. Moreover, a comprehensive study on the layer-dependent nonlinear photonic effect was carried out in MoS_2 mono- and few-layers by CVD growth [6]. SA to TPA transition was confirmed when the thickness changes from few-layer to monolayer. In addition, a spatial self-phase modulation method has been applied to tune the nonlinear refractive index of TMD dispersions [7].

The above-mentioned works have opened up a door towards 2D semiconductor based nonlinear photonics, spectroscopy and relevant photonic devices [8,9].

Acknowledgement.

These works were supported in part by NSFC (No. 61522510) and the External Cooperation Program of BIC, CAS (no. 181231KYSB20130007). J.W. thanks the National 10000-Talent Program for financial support.

References

[1] K. Wang, J. Wang, J. Fan, M. Lotya, A. O'Neill, D. Fox, Y. Feng, X. Zhang, B. Jiang, Q. Zhao, H. Zhang, J.N. Coleman, L. Zhang, W. J. Blau, ACS Nano **7**, 9260(2013).
[2] K. Wang, Y. Feng, C. Chang, J. Zhan, C. Wang, Q. Zhao, J.N. Coleman, L. Zhang, W.J. Blau, J. Wang, Nanoscale **6**, 10530(2014).
[3] N. Dong, Y. Li, Y. Feng, S. Zhang, X. Zhang, C. Chang, J. Fan, L. Zhang, J. Wang, Scientific Reports **5**, 14646 (2015).
[4] X. Zhang, S. Zhang, C. Chang, Y. Feng, Y. Li, N. Dong, K. Wang, L. Zhang, W.J. Blau, J. Wang, Nanoscale, **7**, 2978-2986 (2015).
[5] S. Zhang, N. Dong, N. McEvoy, M. O'Brien, S. Winters, N.C. Berner, C. Yim, X. Zhang, Z. Chen, L. Zhang, G.S. Duesberg, J. Wang, ACS Nano **9**, 7142-7150 (2015).
[6] Y. Li, N. Dong, S. Zhang, X. Zhang, Y. Feng, K. Wang, L. Zhang, J. Wang, Laser & Photonics Reviews **9**, 427-434 (2015).
[7] G. Wang, S. Zhang, X. Zhang, L. Zhang, Y. Cheng, D. Fox, H. Zhang, J.N. Coleman, W.J. Blau, J. Wang, Photonics Research, **3**, A51-A55 (2015).
[8] Y. Li, N. Dong, S. Zhang, K. Wang, L. Zhang, J. Wang, Nanoscale **8**, 1210 (2016).
[9] X. Zhang, Y. Chen, B. Chen, H. Wang, K. Wu, S. Zhang, J. Fan, S. Qi, X. Cui, L. Zhang, J. Wang, Nanoscale **8**, 431 (2016).