

Ultrafast Nonlinear Optical Responses of 2D MoS₂ Nanosheets

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Research on graphene brings not only a new nanomaterial with excellent physical and chemical properties. Most importantly, the material revolutionized the traditional ideology on nanoscience and nanotechnology and opened up a door to a new two-dimensional (2D) nano-system [1]. Following the same vein on the graphene study, researchers have started the exploration of graphene analogue - material comprising stacked atomic or molecular layers [2]. Strong covalent bonds in layer and weak van der Waals interaction between layers allow the graphene analogues forming robust 2D nanostructure. Layered molybdenum disulfide (MoS₂) is one of the typical graphene analogues. Owing to the specific 2D confinement of electron motion and absence of interlayer perturbation, the monolayer MoS₂ shows dramatic improvements in charge carrier mobility and photoluminescence quantum efficiency by factors of 10² and 10⁴, respectively, in comparison with the bulk counterpart [3,4]. The remarkable physical properties of the layered semiconductor nanomaterial inject new opportunities in the field of photonics and optoelectronics.

Whereas the electronic and luminescent properties of 2D MoS₂ nanosheets have been generating much research interest, the ultrafast nonlinear optical (NLO) properties remain largely unexplored. We studied for the first time, to the best of our knowledge, the ultrafast NLO property of MoS₂ nanosheets in liquid-phase dispersions. Employing high-yield exfoliation of MoS₂ in the liquid-phase, a series of dispersions with large populations of monolayer and few-layer MoS₂ were prepared in N-methyl-pyrrolidone (NMP) [5]. As shown in Fig. 1(a) and (b), it is clearly seen that the micro-scale MoS₂ bulk can be effectively exfoliated to 100-nm-scale monolayer and few-layer flakes. UV-Vis-NIR absorption and Raman spectroscopic studies in Fig. 1 (c) and (d) confirmed high quality of the MoS₂ nanosheets. The ultrafast NLO properties were investigated using open-aperture Z-scan technique. All experiments were performed using 100 fs pulses at 800 nm from a mode-locked Ti:Sapphire laser and 6 ns pulses at 532 nm from a Q-switched Nd:YAG laser. Under the excitation of 800 nm fs pulses, the MoS₂ nanosheets exhibited strong saturable absorption (see Fig. 1(e)). Referring to the large bandgap difference between the MoS₂ bulk (1.29eV, ~960nm) and the MoS₂ monolayer (1.90eV, ~650nm), the saturable absorption at 800 nm implies a large number of few-layer MoS₂ nanosheets exist in the NMP dispersions. The monolayer MoS₂ would possess two-photon-absorption at 800 nm fs excitation. For the ns excitation at 532 nm, the MoS₂ nanosheets showed intensity dependent nonlinear extinction behavior - saturable absorption at lower intensity region and induced nonlinear scattering at higher intensity region (see Fig. 1(f)). The variety of the ultrafast NLO responses verifies the 2D MoS₂ a huge potential in the development of nanophotonic devices, such as, mode locker [6] and optical limiter [7]. Part of this work has been submitted for publication elsewhere.

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Figures

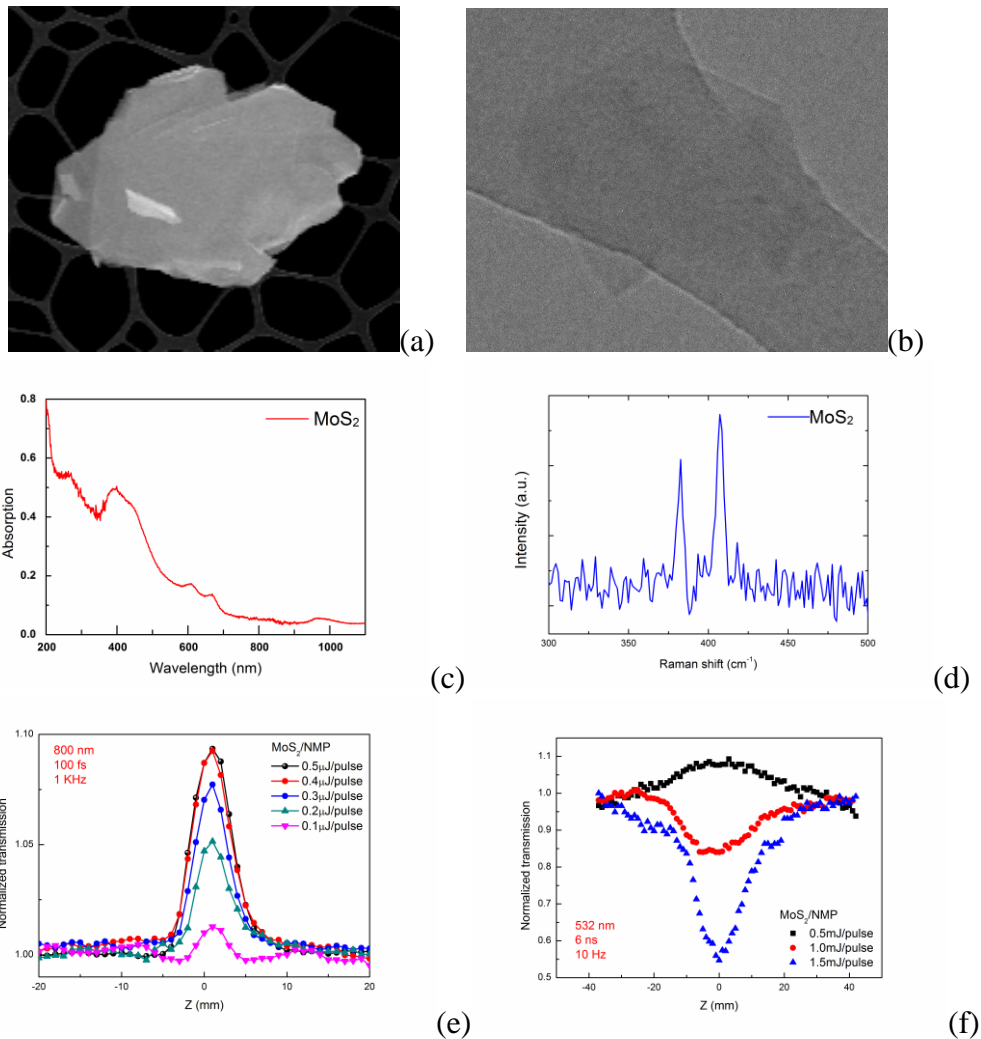


Fig. 1. Ultrafast nonlinear optical responses of 2D MoS₂ nanosheets.