Electronic Supplementary Material (ESI) for Nanoscale.

Tailoring optical properties of atomically-thin WS₂ via ion irradiation

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1. XPS spectra of as-prepared and irradiated WS₂



Figure S1: XPS spectra of as-prepared (S_0) and irradiated (S_1, S_2, S_3) WS₂. W 4f, W 5p states and S 2p states.

2. TEM images of as-prepared and irradiated WS₂



Figure S2: **TEM of as prepared and irraidated WS**₂ **nanosheets**. **a, c,** Low resolution TEM images of flakes of S_0 and S_3 . **b, d,** High-resolution TEM images of S_0 and S_3 .

3. Q-switched lasing based on a Yb:YAG waveguide cavity

A ytterbium doped ytterbium aluminum garnet (Yb:YAG) ceramic waveguide produced by the ultrafast laser writing is used as the gain medium. The detailed information about the waveguide has been reported in Ref. [1]. Mirrors with high reflectivity at 1064 nm and high transmission at 810 nm were coated onto end facts of the waveguide. The pumping laser from a 980 nm continuous solid laser was coupled into the waveguide through a lens (focal length = 20 mm). The output light from the waveguide was collected by a long work distance microscope objective (×20) and detected by a fast photodetector (DET10A/M, Thorlabs, Inc., USA).



Figure S3: Experimental results for the Q-switched pulsed Yb:YAG waveguide lasing. The output power (a), pulse trains (b), pulse duration (c) and modulation depth (d) of the output laser modulated by the as-prepared (S_0) or irradiated (S_3) WS₂ monolayer. MO: Microscope objective.

4. Measurement of the imaginary refractive index of WS₂

The as-prepared (S_0) and irradiated (S_1 , S_2 and S_3) WS₂ were measured by a prism coupler (Metricon 2010). As shown in figure S4a, the WS₂ was pressed tightly onto a precisely characterized rutile prism. The detecting light was incident into the prism and reflected at the interface between the prism and the WS₂. During the measurement, the incident angle of the detecting light was changed. Along with the angular variation, the intensity of the reflected light was measured. Vertical to the basement of the prism, WS₂, MgF₂, the air gap (between WS₂ and prism) and the prism constituted a multilayered structure. In each layer, the electromagnetic fields were written as superposition of positive – and negative – components. Solving the Maxwell equation under the constraint of tangential field continuity, the complex reflective index of the detecting light can be calculated along with the incident angle, following the way described in Ref. [2]. In this work, we focused on the variation of the imaginary refractive index (k) of the WS₂ induced by the ion irradiation.

Figure S4b shows the imaginary refractive index (*k*) of the WS₂ at the wavelength of 1550 nm. With the polarization of the detecting light parallel to the WS₂ film (θ =0°), the imaginary refractive index was observed to be k_{S0} =0.15, k_{S1} =0.2, k_{S2} =0.22 and k_{S3} =0.27, demonstrating the increasing of the optical absorption due to the ion irradiation. Meanwhile, the value of *k* was decreased to 0, with the polarization vertical to the WS₂ film (θ =90°), which indicates the polarization dependent absorption of the WS₂ film.



Figure S4: a, Experimental setup for the prism coupling. b, Imaginary part of S_0 , S_1 , S_2 and S_3 .

Reference

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