Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Direct synthesis of large-scale hierarchical MoS_2 films nanostructured with orthogonally oriented vertically and horizontally aligned layers

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Figure S1

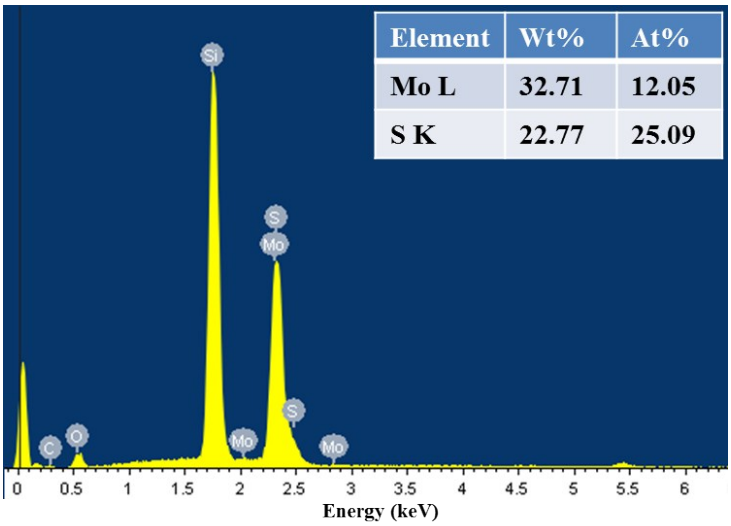

Figure S1. EDS spectrum of the hierarchical MoS_2 nanofilms, which gives a mole ratio of S:Mo of 2.08. The results are the same for both sides of the hierarchical MoS_2 nanofilms.

Figure S2

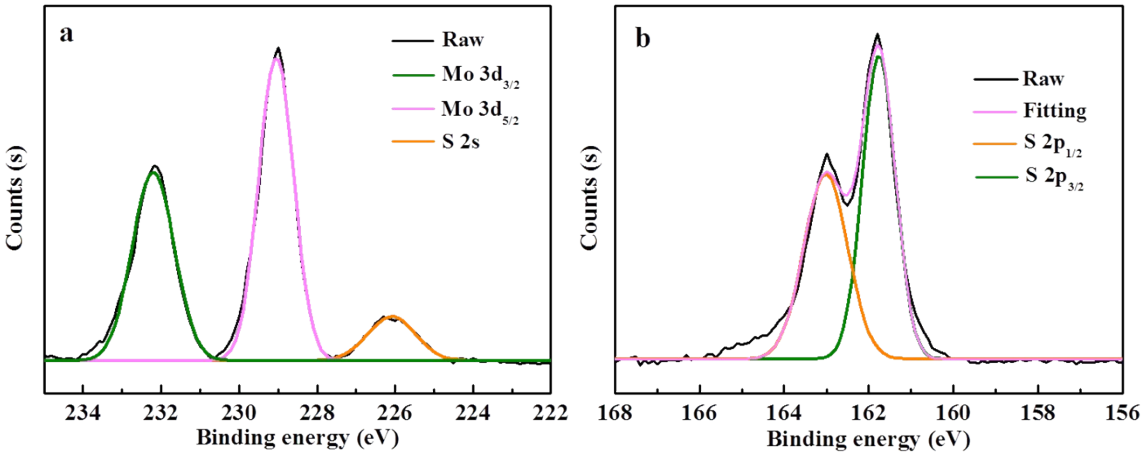

Figure S2. XPS of Mo 3d and (i) S 2p taken from the hierarchical MoS_2 thin films in the NMP-H_2O systems. X-ray photoelectron spectroscopy (XPS) was conducted to analyze the chemical states of Mo and S in the MoS_2 sample. The Mo 3d XPS spectrum shows two sharp peaks at 229.0 and 232.2 eV, which can be indexed to doublet Mo $3d_{5/2}$ and Mo $3d_{3/2}$, respectively. A small peak located at 226.0 eV is assigned to S 2s. The XPS spectrum of S 2p exhibits two peaks at 161.8 and 163.0 eV, corresponding to the S $2p_{3/2}$ and S $2p_{1/2}$ of divalent sulfide ions. The binding energies are consistent with the reported values for MoS_2 crystal.[1-3]

Figure S3

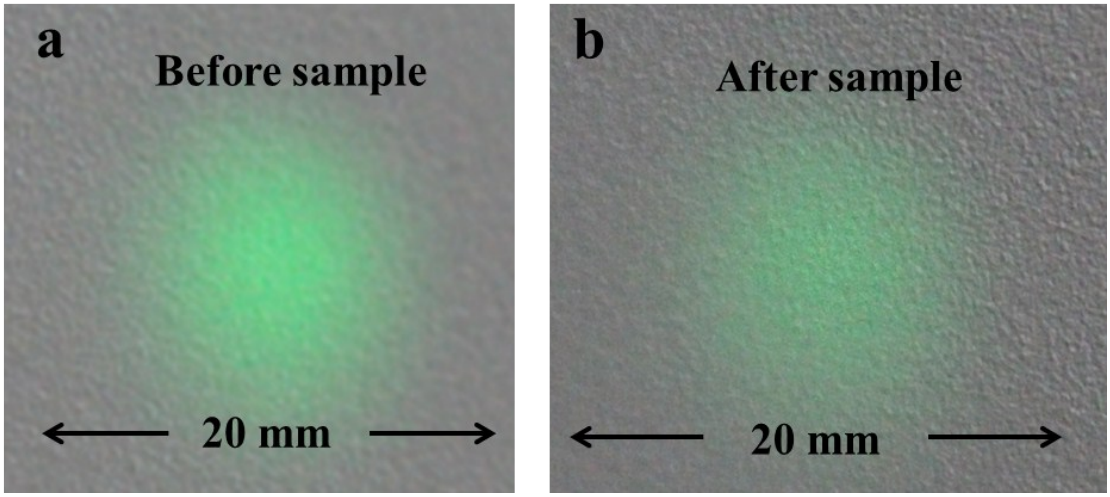

Figure S3. Photograph of a collimated beam (a) before and (b) after passing through the hierarchical MoS_2 thin films (532 nm, CW laser). No significant scattering or distortion is observed for the beam after it passes through the sample, demonstrating the high optical homogeneity of the hierarchical MoS_2 thin films.

Figure S4

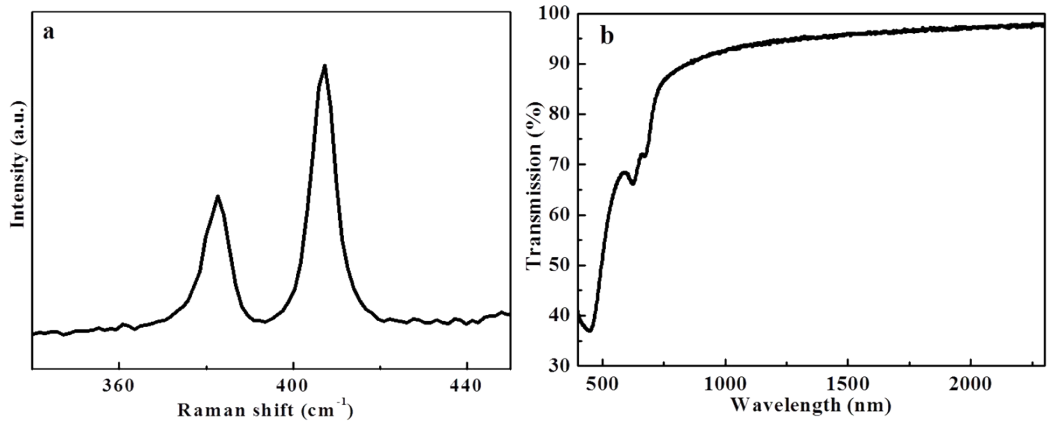

Figure S4. (a)Raman spectrum and (b) Transmission spectrum of the MoS_2 thin films nanostructured with only the horizontally aligned layers prepared with the solvothermal reaction time as 6 h.

Figure S5

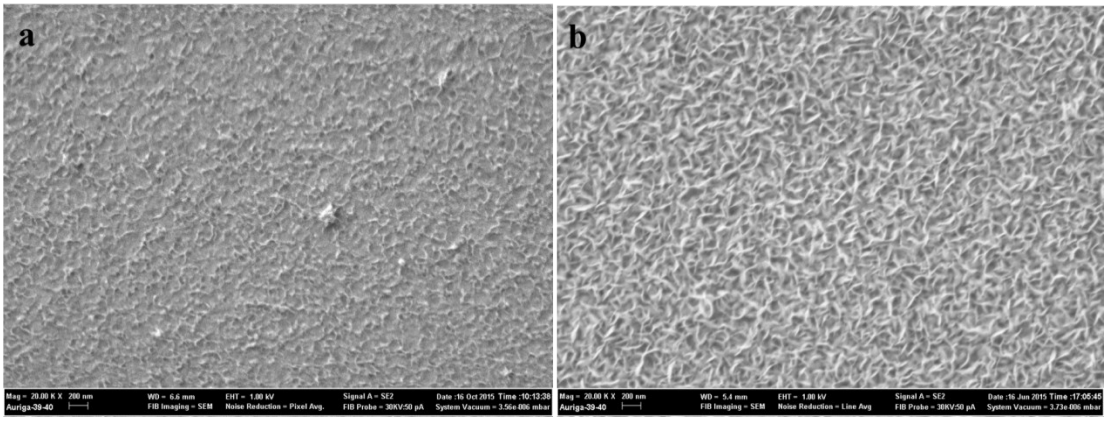

Figure S5. SEM images of the MoS_2 thin films obtained by tuning the solvothermal time to be (a) 8 h and (b) 12 h, respectively.

Figure S6

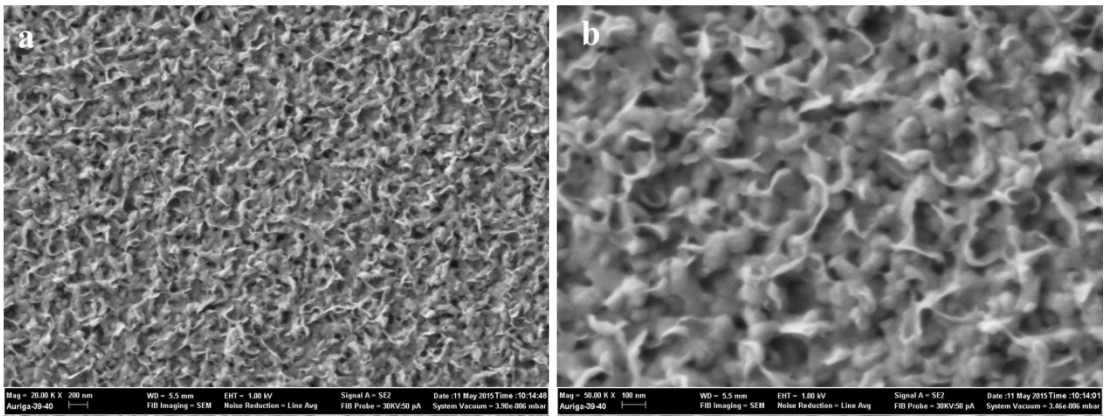

Figure S6 SEM images of the MoS_2 thin films obtained by lowering the concentration of Mo precursor to 0.006 M. (a) Low magnification SEM image, (b) high magnification SEM image.

Figure S7

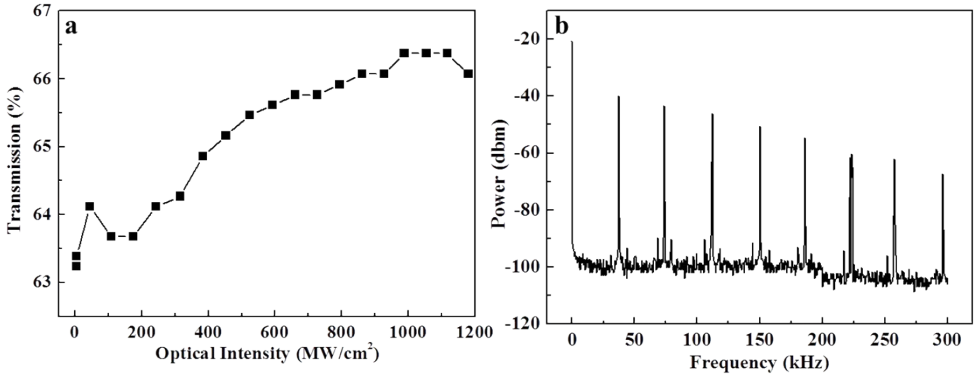

Figure S7. (a) Saturable absorption of the MoS_2-PVA SA, (b) Wide span measurement of the RF spectrum.

Table S1

Table S1 Linear and NLO parameters of the hierarchicalMoS_2 thin films nanostructured with orthogonally oriented layers on quartz substrate with different laser propagation sequences with the film thickness as 160 nm.

Laser propagation sequence	Laser	T (%)	α_0 (cm^{-1})	β ($cm\ GW^{-1}$)	I_s ($GW\ cm^{-2}$)
Back side	1040 nm, 100 Hz	65.0	2.6×10^4	-149±30	78±15
Front side	1040 nm, 100 Hz	65.0	2.6×10^4	-223±30	57±15

Note: L, the film thickness with an error range of ± 10 nm; T, linear transmission at low input energy; α_0, linear absorption coefficient; β, nonlinear absorption coefficient; I_s, saturation intensity.

Table S2

Table S2 Passively Q-switching pulsed lasers in Er-doped fiber laser with few-layer MoS_2 and WS_2 SAs.

Few-Layer MoS_2 Fabrication Method	Integration Platform	Layers in MoS_2 Flakes	λ(nm)	Laser Properties			Ref.
				t	f_{rep}	P(mW)	
SP-LPE (MoS_2)	PVA composite	3-4	1551	3.3 μs	43.4 kHz	5.9	[4]
	PVA composite	~3	1565	5.4 μs	27.0 kHz	1.7	[5]
CVD (MoS_2)	Fiber facet	4-5	1550	1.6 μs	173 kHz	4.7	[6]
ME (MoS_2)	Fiber facet	1-8	1563	3.9 μs	41 kHz	3.5	[7]
SP-LPE (MoS_2)	Tapered fiber	~5	1560	7.5 μs	/	/	[8]
SP-LPE (MoS_2)	PVA composite	~5	1550	3.9 μs	41 kHz	0.7	[9]
SP-LPE (WS_2)	PVA composite	~5	1550	9.9 μs	78 kHz	5.5	[9]
SP-LPE (WS_2)	PVA composite	/	1563	0.71 μs	134 kHz	2.5	[10]
STG (MoS_2)	PVA	4-5	1550	2.2 μs	52 kHz	0.1	Ours

								composite

Note: Where ranges of parameters were reported for Q-switched lasers due to the power-dependent repetition rate and pulse duration, we quote the properties at the maximum power. SP-LPE, solution processed using liquid-phase exfoliation (LPE) method; ME, mechanical exfoliation; STG, solvothermal growth; λ, operating wavelength; t, minimum pulse duration; f rep, highest pulse repetition rate; P, maximum average output power.

References

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