Full Paper
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Supporting Information

Facile fabrication of wafer-scale MoS_2 neat films with enhanced third-order nonlinear optical performance

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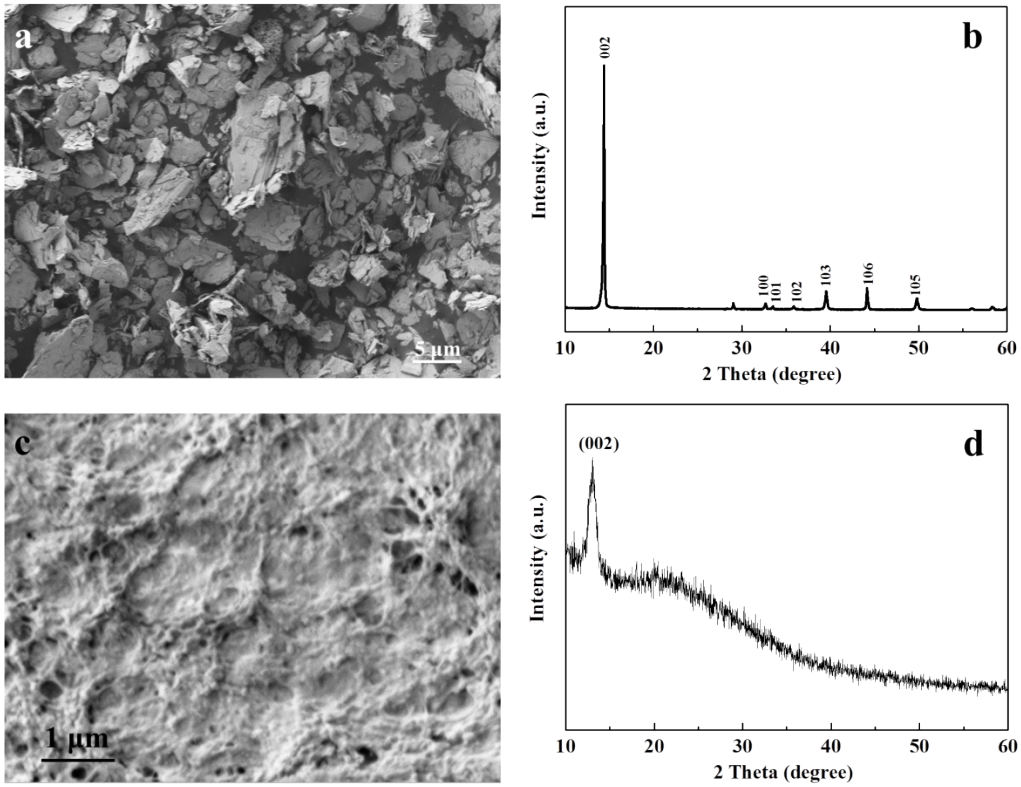

Figure S1. (a) SEM image and (b) XRD pattern of bulk MoS_2 powders. (c) SEM image and (d) XRD pattern of MoS_2 filtered thin film.

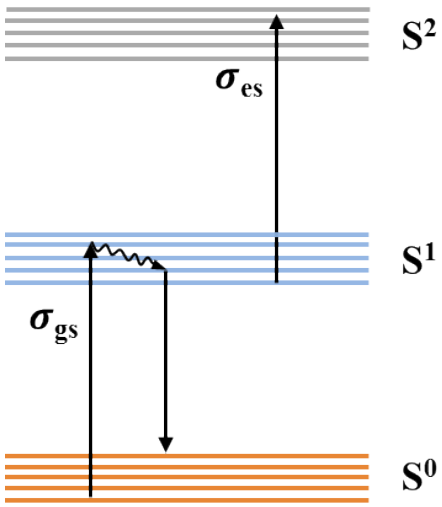

Figure S2. Schematic diagram of a three-level system used for modeling of saturable absorption

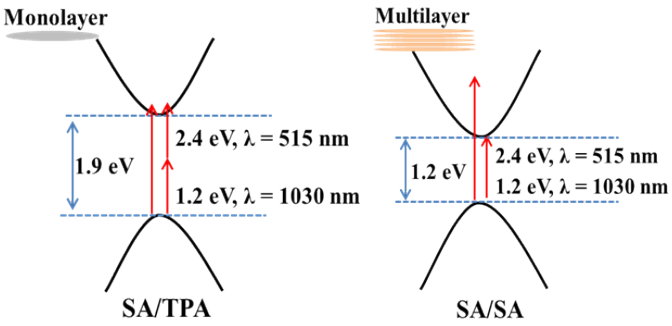

Figure S3. Schematic diagram of NLO mechanism of monolayer and multilayer MoS_2 nanosheets at 515 and 1030 nm. Both monolayer and multilayer MoS_2 nanosheets exhibit saturable absorption (SA) performance at 515 nm with one photon energy of 2.4 eV, while the mechanism at 1030 nm with one photon energy of 1.2 eV is SA for multilayer ones and two-photon absorption (TPA) for monolayer ones

Table S1 Thicknesses of MoS_2 thin films from different MoS_2 dispersions using different membrane pore sizes.

Samples	1500 rpm/1mL			3000 rpm/4mL			6000 rpm/4mL		
Φ_m [nm]	50	100	220	50	100	220	50	100	220
L [nm]	74	63	23	118	111	56	63	60	30

Note: Φ_m, the pore size of nitrocellulose membrane; L, the thickness of transferred MoS_2 films with an error range of ± 5 nm; /1mL, 1 mL of MoS_2 dispersions was diluted into 60 mL for vacuum filtration each time; /4 mL, 4 mL of MoS_2 dispersions was diluted into 60 mL for vacuum filtration each time.

Table S2 The average surface roughness for MoS_2 thin films with different film thicknesses from different MoS_2 dispersions using different membrane pore sizes.

Samples	*r*/rpm[a]:	1500	3000	6000	Φ_m/nm[b]:	50	100	220	6000 rpm[c]		
L [nm]		63	47	45		60	47	56	45	63	70
R [nm]		13	14	15		11	12	9	12	14	13

Note: *L*, the thickness of transferred MoS_2 films with an error range of ± 5 nm; *R*, the mean surface roughness of the films obtained from AFM with an error range of ± 2 nm; a, the films fabricated with different centrifugation rates (1500, 3000 and 6000 rpm) with membrane pore size of 100 nm; b, the films with different membrane pore sizes (50, 100, and 220 nm) from dispersions with centrifugation rate as 3000 rpm; c, the films with different thicknesses from dispersions with centrifugation rate of 6000 rpm.

Table S3 Linear and NLO parameters of transferred MoS_2 neat film in compared to those of MoS_2 dispersions in cyclohexyl pyrrolidinone (CHP) and graphene filtered film.

Sample	Laser	*L*	*T* [%]	α_0 [cm^{-1}]	*n*	α_{SA} [cm GW^{-1}]	I_s [GW cm^{-2}]	Im $\chi^{(3)}$ [esu]	FOM [esu cm]
MoS_2 film	515 nm, 100 Hz	90 nm	65.0	4.7×10^4	6.8 [1]	-2.1×10^4	2.2	-1.6×10^{-7}	3.3×10^{-12}
	1030 nm, 1000 Hz	90 nm	87.0	1.5×10^4	4.4	-82.9	171	-5.3×10^{-10}	3.4×10^{-14}
MoS_2 in	515 nm,	1	7.9	25.34	1.5	-0.36	58	$-1.3\times10^{-}$	$5.1\times10^{-}$

	1000 Hz	mm						^{13}	^{15}
CHP [2]	1030 nm, 1000 Hz	1 mm	30.9	11.75	1.5	-9.2×10^{-2}	114	-6.7×10^{-14}	5.7×10^{-15}
Graphene film	515 nm, 100 Hz	120 nm	64.6	3.6×10^{4} [3]	2.6	-2.0×10^{4}	1.5	-2.2×10^{-8}	6.1×10^{-13}
	1030 nm, 100 Hz	110 nm	83.0	1.7×10^{4}	2.6	-76.4	192	-1.7×10^{-10}	1.0×10^{-14}

Note: L, the path length of the samples, for film is the film thickness with an error range of ± 5 nm, for dispersion is the path length of quartz cuvette; T, linear transmission at low input energy; α_0, linear absorption coefficient; n, refractive index; α_{SA}, nonlinear absorption coefficient; I_s, saturation intensity; Im $\chi^{(3)}$, the imaginary third-order susceptibility, $Im\,\chi^{(3)} = \left[\frac{10^{-7}c\lambda n^2}{96\pi^2}\right]\alpha_{SA}$; [4] FOM, figure of merit (FOM) for the third-order optical nonlinearity, $FOM = \left| Im\,\chi^{(3)}/\alpha_0 \right|$. [4] MoS_2 dispersions in CHP were obtained by liquid exfoliation method. [2]

Table S4 NLO parameters of transferred MoS_2 neat film in compared to that of graphene filtered film.

Sample	Laser	T [%]	σ_{gs} [cm^2]	σ_{es} [cm^2]	σ_{gs}/σ_{es} [cm^2]	N [cm^3]
MoS_2 film	515 nm	65.0	4.7×10^{-15}	2.0×10^{-15}	0.7	1.0×10^{19}
	1030 nm	87.0	2.9×10^{-17}	1.5×10^{-17}	0.5	5.3×10^{20}
Graphene film	515 nm	64.6	3.3×10^{-15}	6.4×10^{-16}	0.2	1.0×10^{19}
	1030 nm	83.0	4.0×10^{-17}	2.8×10^{-17}	0.7	3.8×10^{20}

Note: σ_{gs}, ground-state absorption cross section; σ_{es}, excited-state absorption cross section; N, the absorber's effective density.

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