Supplementary Information

Preparation of Monolayer MoS$_2$ Quantum Dots using Temporally Shaped Femtosecond Laser Ablation of Bulk MoS$_2$ Targets in Water

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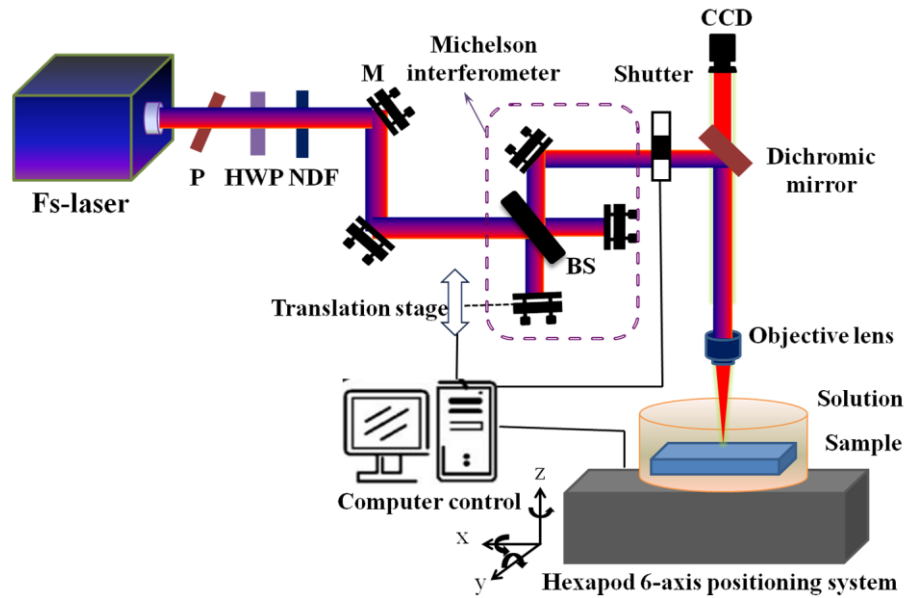
Figure S1. Schematic of the experimental setup for two-subpulse generation. P: polarizer; HWP: half-wave plate; NDF: neutral density filters; M: mirror; BS: beam splitter; CCD: charge coupled device.
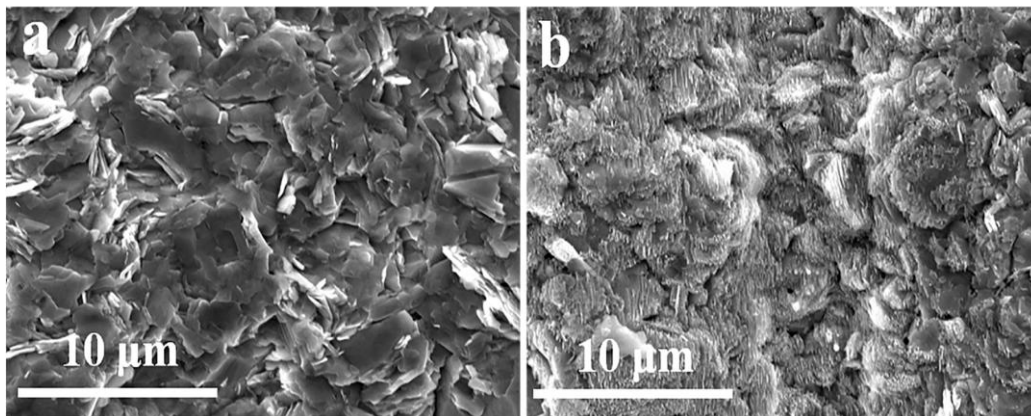

Figure S2. SEM images of the surface of original bulk MoS_2 target (a) before and (b) after femtosecond-laser temporally shaped two-subpulse train ablation in water.
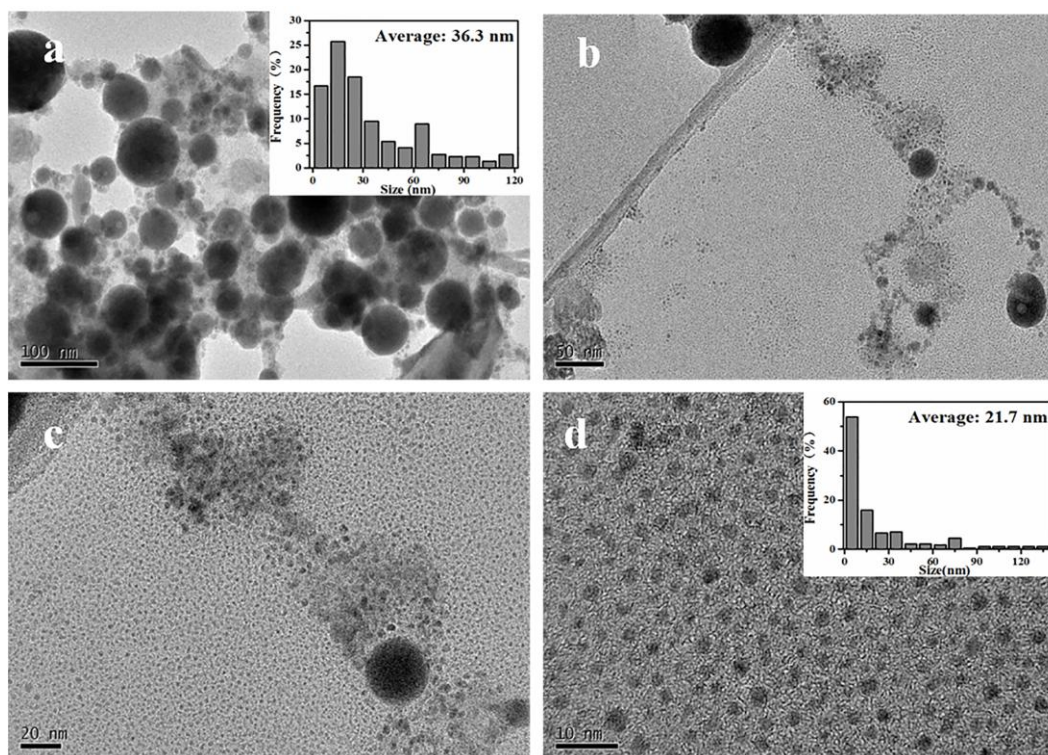
**Figure S3.** The TEM images corresponding to the morphology of MoS$_2$ nanomaterials in the aqueous solutions before centrifugation: (a) obtained by femtosecond laser single pulse ablation; (b), (c), and (d) obtained by temporally shaped femtosecond laser two-subpulse train ablation. The insert image of a and d show the size distribution. The scar bars are 100 nm, 50 nm, 20 nm, and 10 nm, respectively.
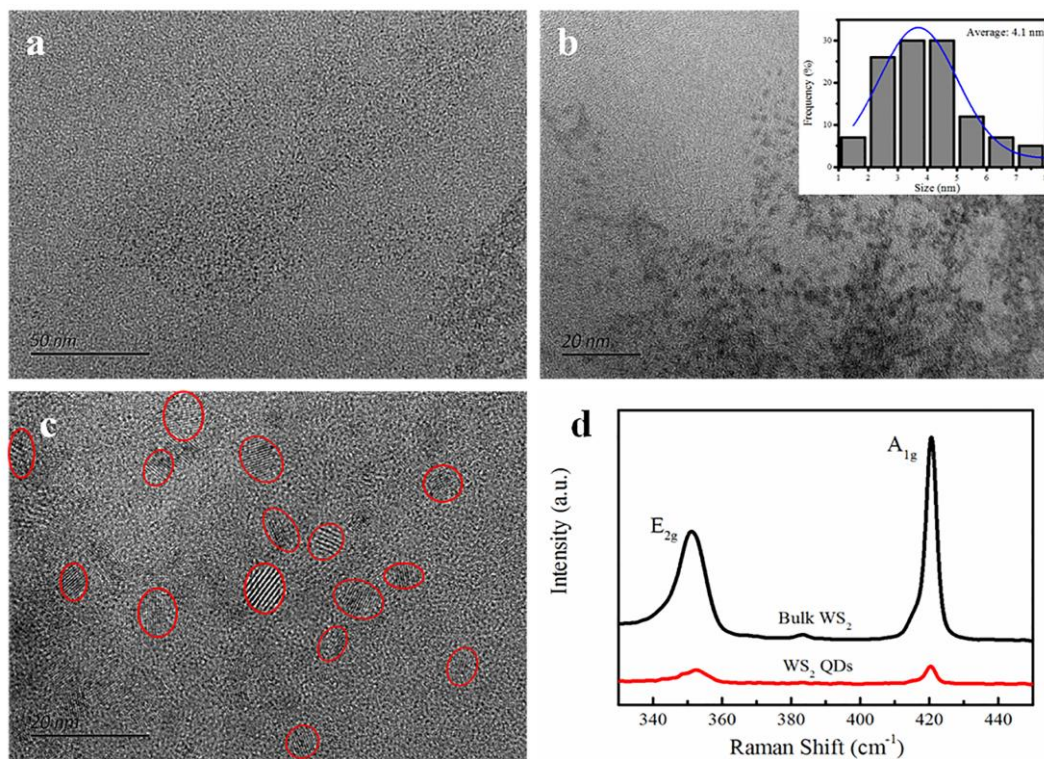
Figure S4. (a-c) TEM and HRTEM images of the as-prepared WS_2 QDs, the scale bars are 50 nm, 20 nm, and 20 nm, respectively. The inset in (b) is the size distribution of the WS_2 QDs. (d) The Raman spectrum of the bulk WS_2 and WS_2 QDs.
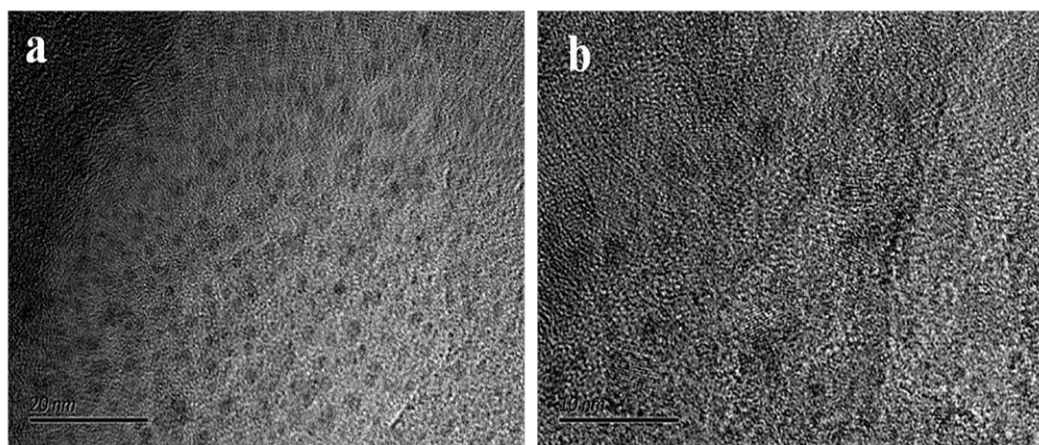
Figure S5. The TEM image of the as-prepared GQDs. The scale bars are 20nm and 10 nm, respectively.
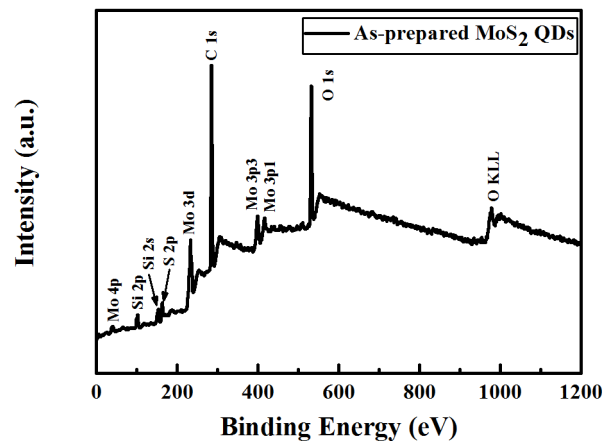
Figure S6. The full spectrum of XPS survey of the as-prepared MoS_2 QDs.
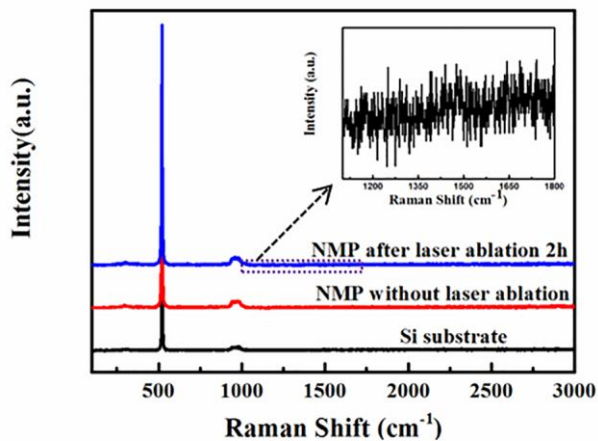
Figure S7. The Raman spectra of the Si substrate, NMP solution without laser ablation, and NMP solution after laser ablation for 2h, respectively. And the inset image is the local magnification of the spectra of NMP solution after laser ablation for 2h.
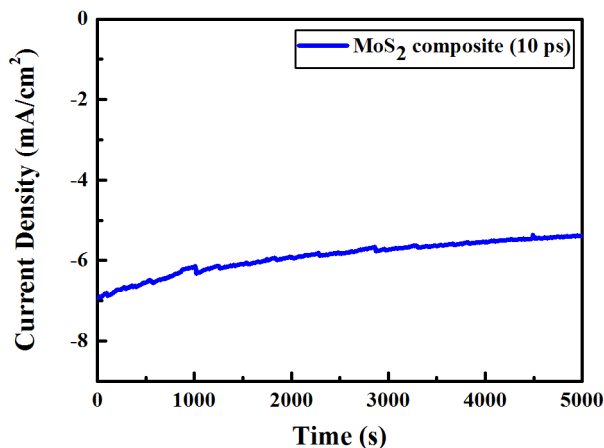
Figure S8. The durability test of MoS_2 composites (composite of MoS_2 nanosheets, nanoparticles, and QDs obtained by temporally shaped femtosecond laser before centrifugation) with an applied voltage of -0.3 V vs RHE over 5000 seconds in 0.5 M H_2SO_4.

Table S1. Comparison of HERs performance with the MoS_2-based HERs catalysts prepared by other typical synthesis methods.

Synthesis methods	Catalysts	Onset potential (V)	Overpotential (V)	Tafel slope (mV dec^{-1})	Refs.
Precursors reaction	Defect-free MoS_2 NSs	-0.18	/	87	56
Sonication combined with centrifugation	MoS_2 NPs	-0.16	-0.2 (0.4 mA cm^{-2})	82	S2
Liquid exfoliation	MoS_2 QDs on NSs	-0.19	-0.4 (120 mA cm^{-2})	74	20
Sonication combined with solvothermal	MoS_2 QDs	-0.12	/	69	18
Ultrasonication combined with centrifugation	MoS_2 NPs	-0.09	-0.15 (0.92 mA cm^{-2})	69	45

Ionic liquid assisted grinding exfoliation	MoS_2 NDs	-0.09	-0.248 (10 mA cm^{-2})	61	9
Electrochemical etching	MoS_2 QDs	-0.21	/	60	22
Hydrothermal	MoS_2 QDs	-0.16	-0.4 (39 mA cm^{-2})	59	21
Precursors reaction	Defect-rich MoS_2 NSs	-0.12	-0.12 (13 mA cm^{-2})	50	56
Chemical exfoliation	MoS_2 NSs on graphite	/	-0.187 (10 mA cm^{-2})	43	53
Solvothermal	MoS_2 NPs on RGO	-0.1	/	41	S1
Temporally shaped femtosecond-laser ablation	MoS_2 composites	-0.14	-0.4 (36 mA cm^{-2})	66	This work

NSs: Nanosheets

NPs: Nanoparticles

NDs: Nanodots

RGO: Reduction graphene oxide

References

S1. Y. Li, H. Wang, L. Xie, Y. Liang, G. Hong and H. Dai, *J. Am. Chem. Soc.* **2011**, *133*, 7296.

S2. T. Y. Wang, D. L. Gao, J. Q. Zhuo, Z. W. Zhu, P. Papakonstantinou, Y. Li, M. X. Li, *Chem.–Eur. J.* **2013**, *19*, 11939.