Supporting Information:

Seeded Preparation of Ultrathin FeS_2 Nanosheets from Fe_3O_4 Nanoparticles

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Experimental Section

Materials. Ferric acetylacetonate ($Fe(acac)_3$, 99.9%) and ammonium thiomolybdate ($(NH_4)_2MoS_4$ were purchased from Aldrich. Octadecylamine (ODA, 90%), diphenyl ether (DE, 99%), and oleylamine (OLA, 90%) were obtained from Aladdin. Sulfur powder (S), acetone, *N, N*-Dimethylformamide (DMF) and chloroform were all commercially available products and used as received without further purification.

Preparation of Fe_3O_4 nanoparticles (NPs). $Fe(acac)_3$ (176 mg, 0.5 mmol), 7.5 mL ODA, and 2.5 mL OLA were mixed in a four-necked flask at 120 °C and degassed under vacuum for 1 h to produce Fe precursor solution. After further heating and annealing treatment at 200 °C for 1 h, the uniform Fe_3O_4 NPs with the average diameter of 3.9 ± 0.6 nm were obtained. As heating and annealing treatment at 200 °C for 10 min, heterogeneous Fe_3O_4 NPs with the diameter of 3.8 ± 1.6 nm were obtained.

Preparation and purification of ultrathin FeS_2 nanosheets. S (144 mg, 4.5mmol) and 5 mL DE were mixed in another three-necked flask at 70 °C and degassed for 1 h to produce the S solution. Subsequently, the S solution was rapidly injected into the Fe_3O_4 NPs ODA/OLA solution and maintained at 200 °C for 60 min to produce ultrathin FeS_2 naonsheets. After cooled down to room temperature, 1 mL FeS_2 naonsheets solution were washed and precipitated through the addition of 1 mL chloroform and 2 mL acetone for two times. Separated by centrifugation, the precipitates were collected and dispersed in 1 mL chloroform. In control experiment, the products were prepared by the same experimental procedure, except injecting the S solution at 220 °C (Figure S10) and 240 °C (Figure S11). To reveal the influence of coordinative solvent, the products were prepared with the ODA/OLA volume ratio of 1/3, while other experimental variables were fixed (Figure S12).

Preparation and purification of 8.3 nm FeS_2 NPs. Pyrite NPs were prepared by

injecting S solution (96 mg S in 5 mL DE) into Fe solution (100 mg $FeCl_2 \cdot 6H_2O$ in 10 mL ODA) at 220 ^oC and stirring for 3 h. 1 mL resultant solution were washed and precipitated through the addition of 1 mL chloroform and 2 mL acetone for two times at room temperature. Separated by centrifugation, the precipitates were collected and dispersed in 1 mL chloroform.

Preparation of ultrathin *2H*-MoS_2 nanosheets. Edge-rich *2H*-MoS_2 ultrathin nanosheets were prepared according to the previous publication (*ACS Appl. Mater. Interfaces* 2013, 5, 12794-12798). 5 mg $(NH_4)_2MoS_4$ was foremost dispersed in the mixture of 2 mL DMF and 1 mL H_2O by ultra-sonication for 30 min. The mixture was then transferred into an autoclave, and maintained at 210 ^oC for 18 h. After cooled down to room temperature, the products were collected and treated by water and ethanol three times, and finally dried at 60 ^oC for 24 h.

Characterization. UV-visible (UV-vis) absorption spectra were measured using a 3600 UV-VIS-near infrared (NIR) spectrophotometer (Shimadzu). Transmission electron microscopy (TEM) and selected area electron diffraction (SAED) were recorded using a H-800 electron microscope (Hitachi) at an acceleration voltage of 200 kV with a charged coupled device camera. High-resolution TEM (HRTEM) images were recorded using a JEM-2100F electron microscope (Jeol) at 200 kV. An energy-dispersive X-ray spectroscopy (EDX) detector coupled with a XL30 ESEM FEG scanning electron microscope (FEI) was used for elemental analysis. Inductively coupled plasma (ICP) was performed with an OPTIMA 3300 DV analyzer (PerkinElmer). X-ray diffraction (XRD) was carried out on a X-ray diffractometer (Rigaku) using CuK radiation (λ =1.5418 Å).

Electrochemistry. A conventional three-electrode cell was employed to perform the electrochemical test. The working electrode was a 5 mm diameter glassy carbon that was carefully polished and ultrasonically rinsed in absolute ethanol before use. The counter electrode was a platinum wire, and the reference electrode was an aqueous

Ag/AgCl electrode in saturated KCl solution. All of the potentials were referred to NHE by adding +0.197 V to the potential vs Ag/AgCl electrode. 0.02 mg Catalysts, including FeS_2 NPs, nanoplates, nanosheets, and *2H*-MoS_2 nanosheets were deposited on the electrode surface, respectively. And then dried in air and left for 12 h at 100 °C in an oven. A 0.025 mol/L phosphate buffer (pH 7.0) was prepared and used as the supporting electrolyte.

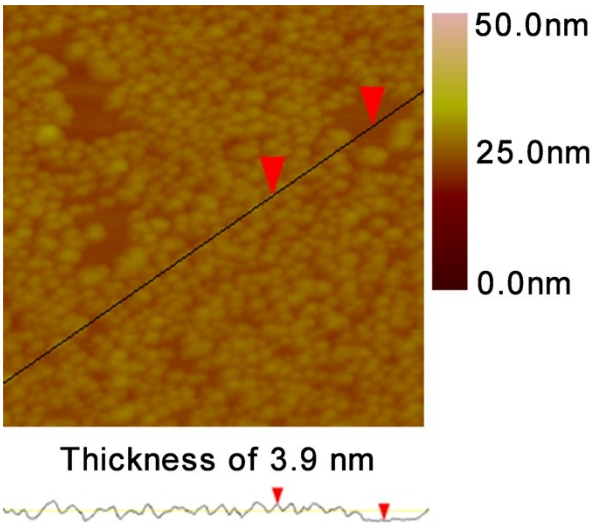

Figure S1. AFM image of Fe_3O_4 seeds. The average diameter is 3.9 nm.

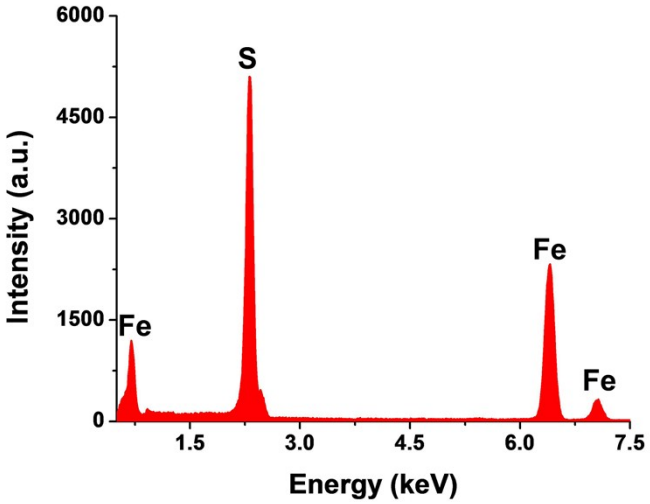

Figure S2. The composition of as-prepared FeS_2 nanosheets is characterized by EDX, which shows the Fe/S molar ratio of 1/2.

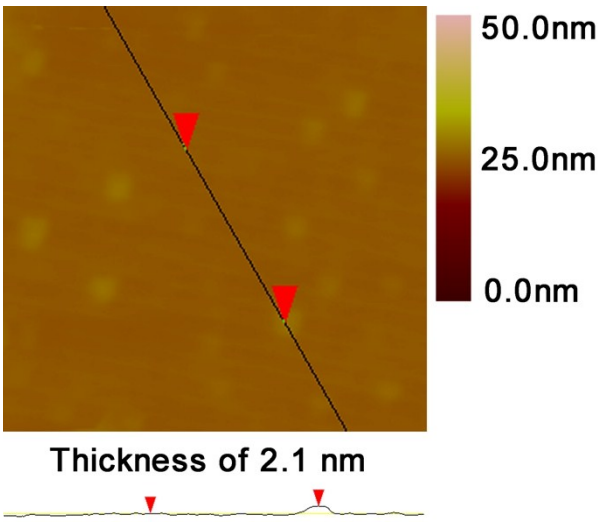

Figure S3. AFM image of FeS_2 nanosheets, those are prepared at 200 ^oC for 1 h. The thickness of nanosheets is 2.1 nm.

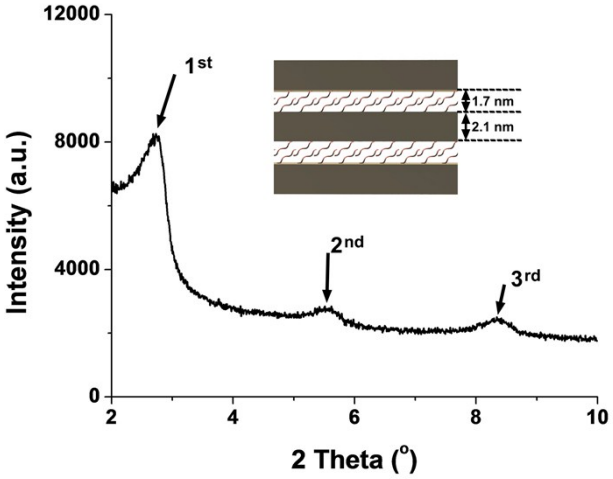

Figure S4. The small-angle XRD pattern of ultrathin FeS_2 nanosheets, which reveals a set of distinct peaks corresponding to the spacing of 3.8 nm, which consists with the sum of FeS_2 nanosheets (2.1 nm) and the thickness of ligand bilayer (1.7 nm).

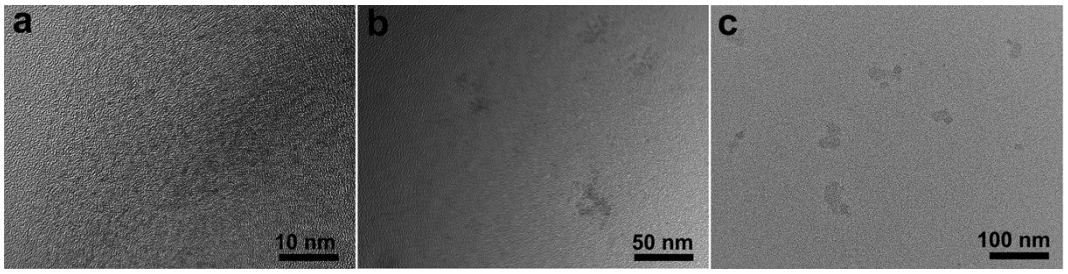

Figure S5. TEM temporal evolution of FeS_2 nanosheets after injection of S solution for 5 (a), 10 (b), and 20 (c) min, which exhibits the tendency of the aggregation of small FeS_2 nuclei.

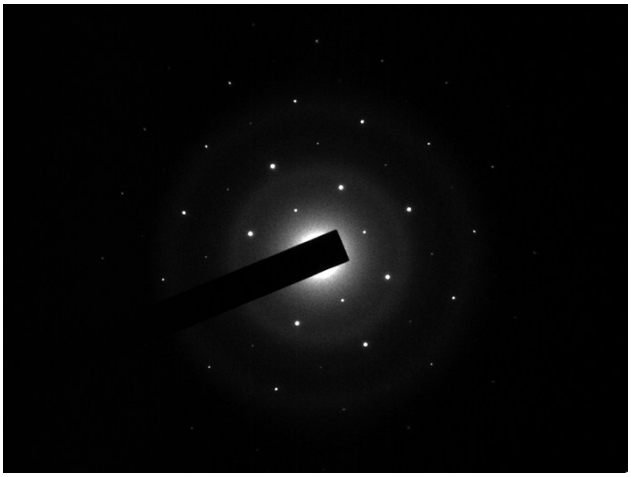

Figure S6. The SAED pattern of one FeS_2 nanosheet, which presents the single-crystalline feature.

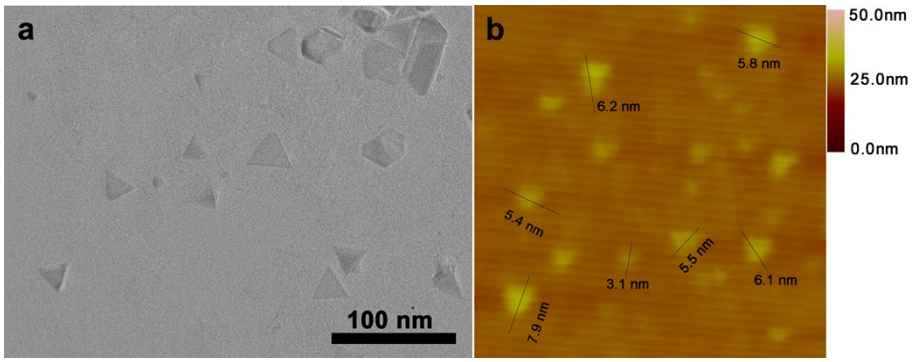

Figure S7. TEM (a) and AFM (b) images of the inhomogeneous FeS_2 nanoplates with different thickness. The average thickness of the nanoplates is 5.7 nm.

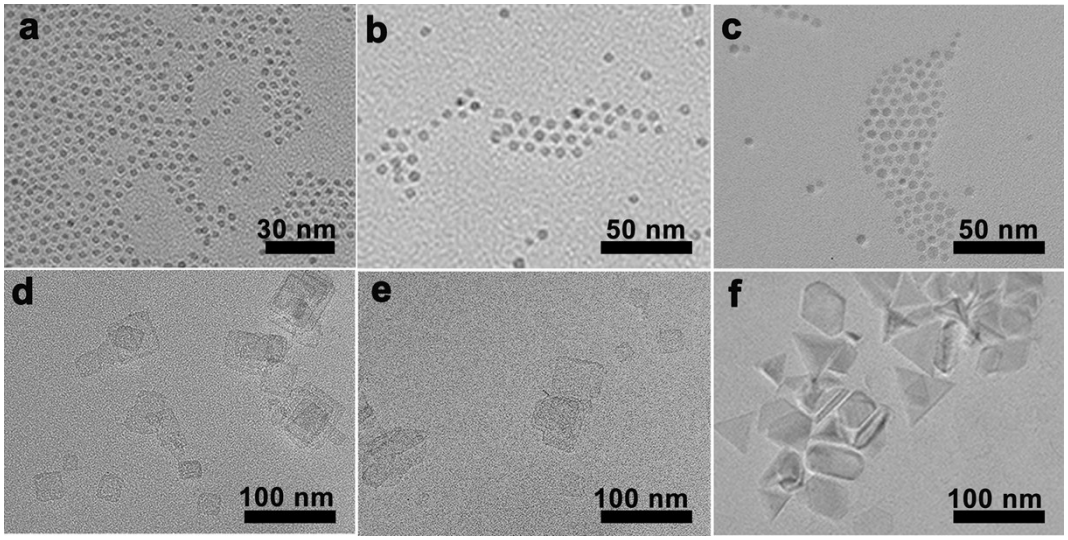

Figure S8. TEM images of Fe_3O_4 seeds with the average diameter of 3 nm (a), 5 nm (b), and the mixed seeds (c), which produce the corresponding FeS_2 nanostructures of ultrathin sheets (d-e) and inhomogeneous plates (f).

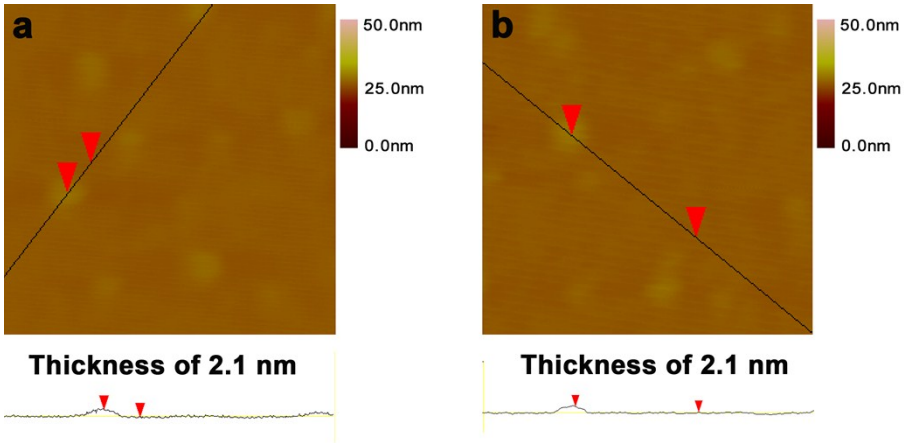

Figure S9. AFM images of the FeS_2 nanosheets that are produced from 3 nm (a) and 5 nm (b) Fe_3O_4 seeds. The thickness is 2.1 nm. The corresponding TEM images are respectively shown in Figure S8d and e.

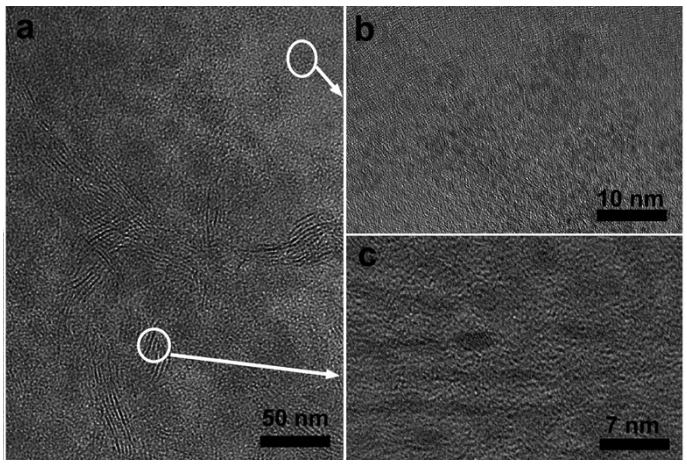

Figure S10. TEM images of the products that are obtained as injection of S solution at 220 °C and annealing at 200 °C for 60 min. There are some big particles loaded on the nanosheets, which leads from the nonuniform FeS_2 nuclei.

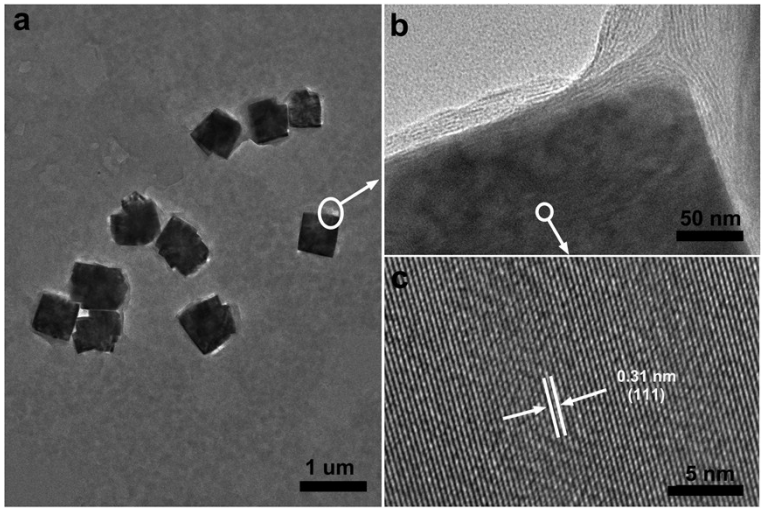

Figure S11. TEM (a, b) and HRTEM (c) images of the products that are prepared as injection of S solution at 240 °C and annealing at 200 °C for 60 min. Besides nanosheets, big cubic crystals are also produced. HRTEM shows the lattice fringe spacing of 0.31 nm, corresponding to the (111) plane of cubic FeS_2.

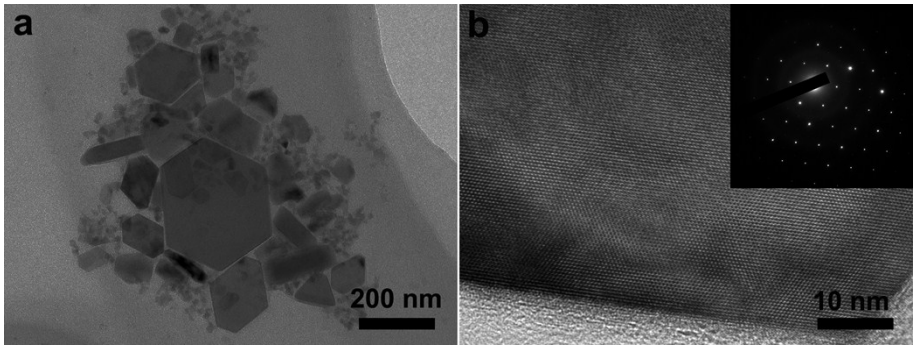

Figure S12. TEM (a) and HRTEM (b) images of the products that are prepared with the ODA/OLA volume ratio of 1/3. Inset in (b): the corresponding SAED pattern.

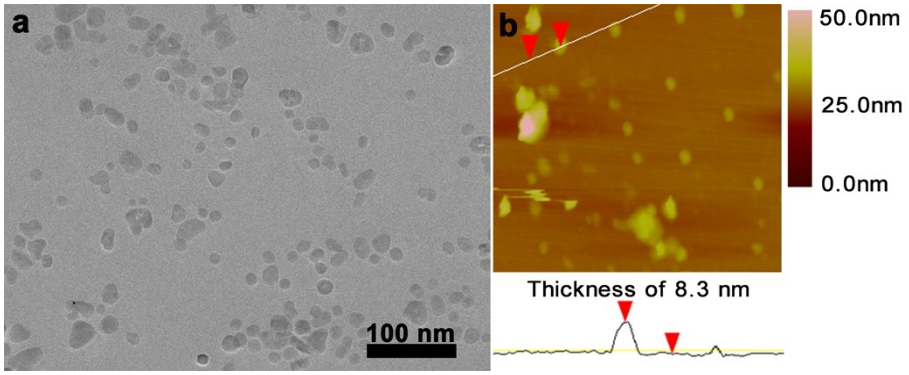

Figure S13. TEM (a) and AFM (b) images of big FeS_2 NPs. The average diameter of the NPs is 8.3 nm.

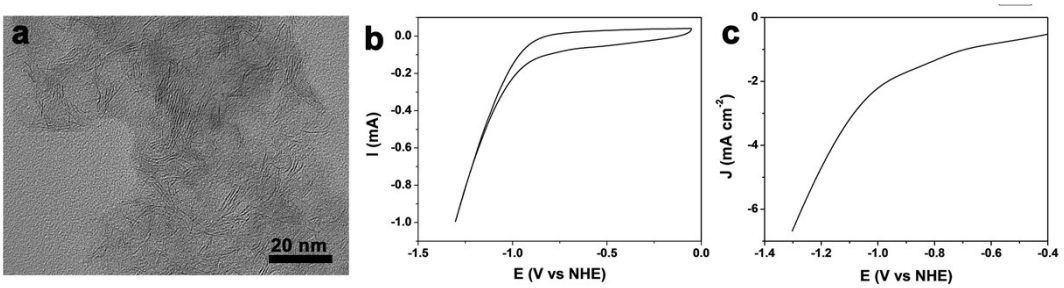

Figure S14. TEM image (a), cyclic voltammogram (b) and linear sweep voltammetry (c) curves of the edge-rich *2H*-MoS_2 ultrathin nanosheets. At the overpotential of 1.2 V (vs NHE), the current density is 4.7 mA/cm^2.

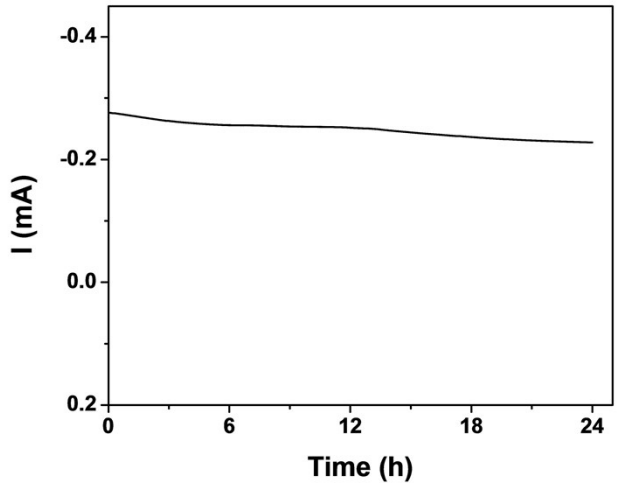

Figure S15. Current *vs* time profile for a 24 h constant potential (-1.0 V *vs* NHE), which indicates the good stability of the FeS_2 nanosheets.