

Electronic Supplementary Information

Facile and efficient exfoliation of inorganic layered materials using liquid alkali metals alloy

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S1. Experimental Section

Intercalation and exfoliation of the MoS₂, WS₂ and BN:

Na (1 g, Aladdin) and K (1 g, Aladdin) and 200 ml 1,2-dimethoxyethane (DME) are bubbled with argon for 5 min and stirred for 7 h at room temperature. We can obtain liquid NaK alloy dispersed in a blue solution. MoS₂ powder (1.2 g) is added to this blue solution and the mixture is further stirred for 24-48 h. A dark dispersion of intercalated MoS₂ was made. After reaction, the product was centrifuged at 8000 rpm for 30 min to remove the DME. The procedures are similar for the NaLi alloy and other single metals (Li, Na, K). Intercalation process for WS₂ is similar except that 1.5 g of WS₂ is used with the same amount of reagent above. Distilled water (200 ml) is added to the intercalated sample to form a homogeneous suspension that was put through dialysis for several days to remove any remaining alkali metal species. The resulting homogeneous dispersion was tested to be stable for several months.

Characterization:

AFM measurements were performed with a SPI 4000 system in tapping mode. The samples of water suspension were deposited on a SiO₂/Si substrate by spin-coating, dried by evaporation, and the measurements performed in air at ambient temperature and pressure. TEM images were obtained using a JEM-1200EX. The TEM samples were prepared by drying a droplet of the graphene suspension on holey carbon grids. SEM imaging was performed using a JSM 7401F. The SEM samples were prepared by depositing materials onto a silicon substrate by drop-drying. Raman spectroscopy was performed on a Renishaw RM2000 microscopic confocal Raman spectrometer (Gloucestershire, United Kingdom) using green (514 nm) laser excitation. Scans were taken on an extended range (1,000–3,500 cm⁻¹). A sample was sonicated in ethanol and drops applied to a glass slide for observation. Raman spectra of solid samples were taken by depositing the graphite on a glass slide. X-ray diffraction data were collected on a Bruker D8-Advance using Cu-K α radiation ($\lambda=1.5418$ Å).

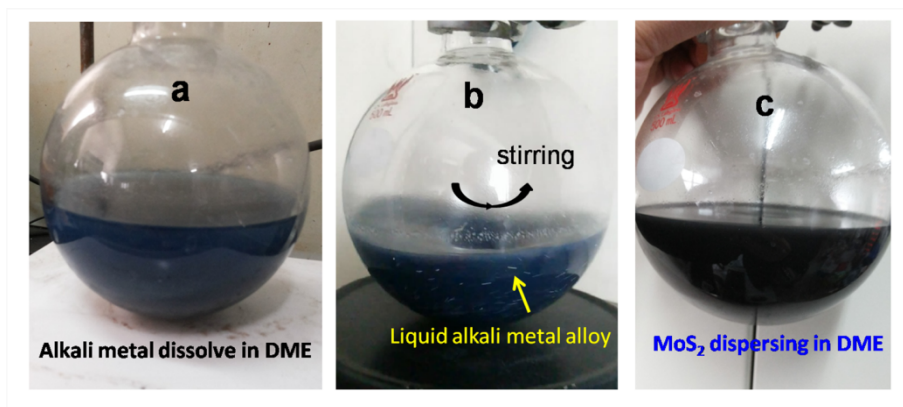


Figure S1 Alkali metal mixture was dissolved in DME to obtain a blue solution (a) and liquid alkali metal alloy (b). MoS₂ was then dispersed in DME to obtain a dark solution (c).

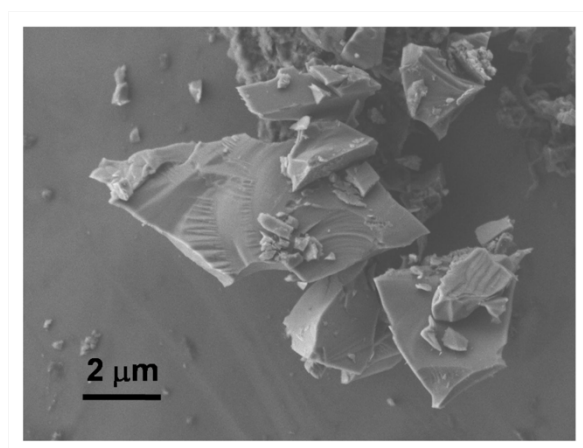


Figure S2 SEM images of bulk MoS₂.

S2. Dispersion of the BN and UV-vis spectra of MoS₂, WS₂ and BN

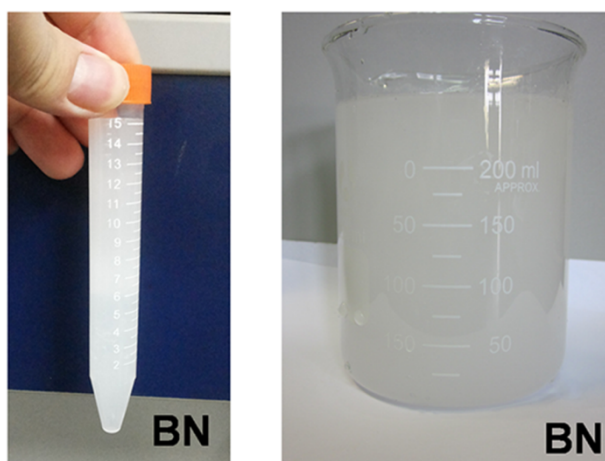


Figure S3 photo images of dispersions of the NaK-exfoliated BN in water.

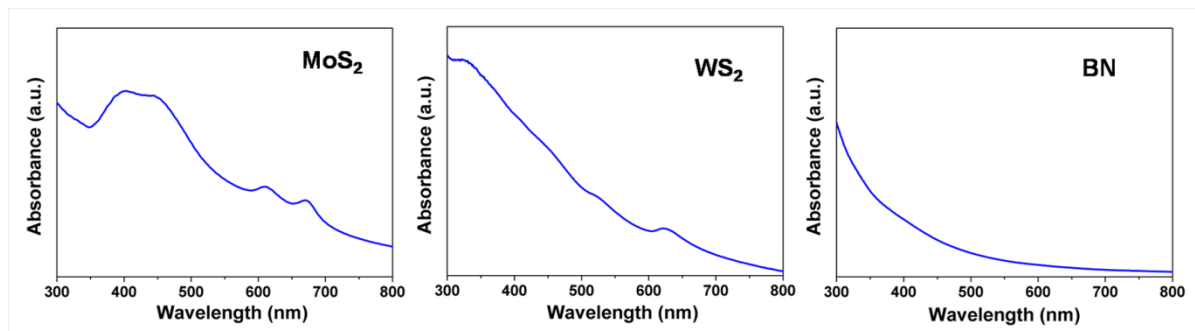


Figure S4 UV-vis spectra of MoS₂, WS₂ and BN dispersions in water.

S3. TEM and EDS of MoS₂ nanosheets

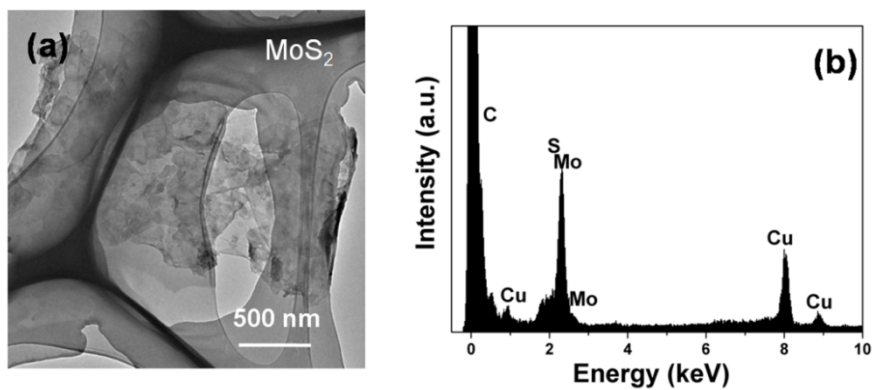


Figure S5 TEM (a) and the EDS spectrum (b) of MoS₂ nanosheets. Elemental analysis of exfoliated MoS₂ sample using the EDS spectrum of showing the presence of Mo and S.

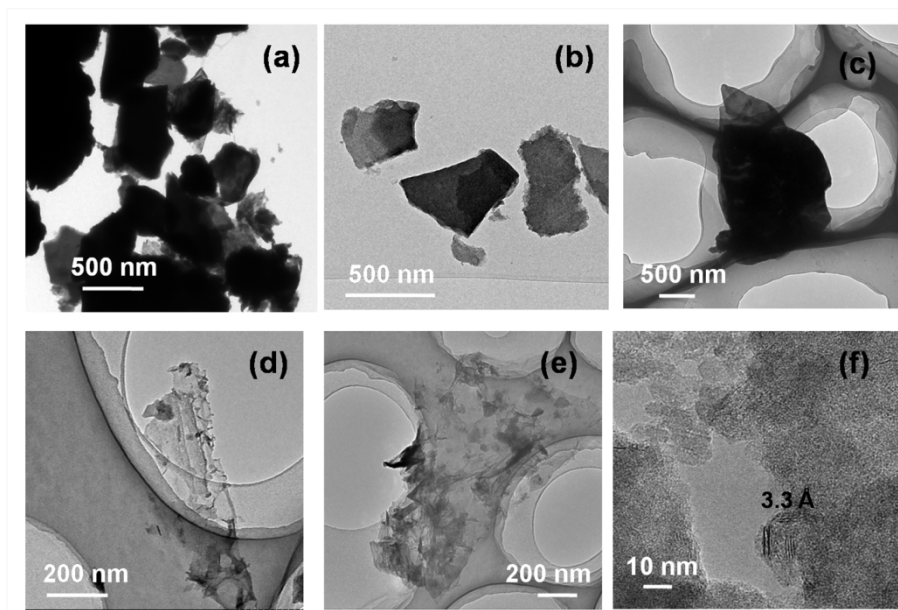


Figure S6 TEM images of MoS₂ nanosheets exfoliated by single alkali metal Li (a), Na (b), K (c), and alkali metal alloy NaLi (d) and NaK (e), and HTEM image of MoS₂ exfoliated by NaK (f) .

S4. XRD of MoS₂ before and after exfoliation

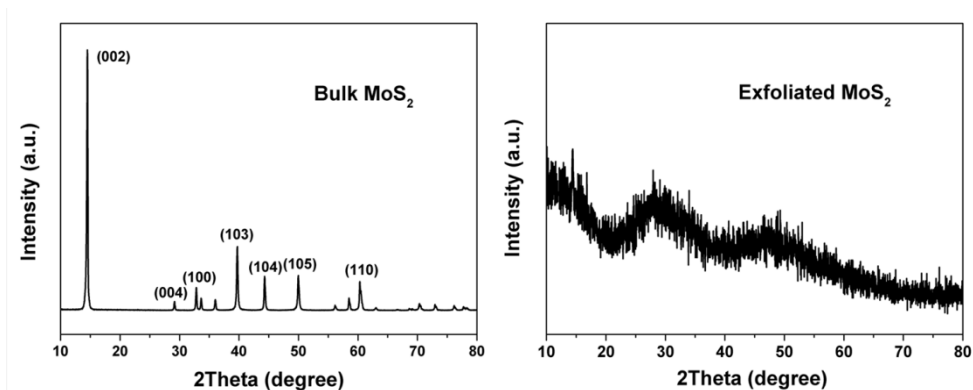


Figure S7 XRD of MoS₂ before and after exfoliation.

S5. MoS₂ and WS₂ film fabrication and EDS and Raman Characterization

(1) Fabrication of MoS₂ thin films on PET substrate and WS₂ film on PVDF film

A dilute MoS₂ suspension was vacuum filtered using a mixed cellulose ester membrane with 20 nm pores (Millipore). The MoS₂ film was allowed to dry and adhered to the substrate at room temperature overnight under a 5.0 kg weight. The

weight was removed and the membrane dissolved in acetone to leave the GO thin film on the substrate. The MoS₂ film was then carefully transferred onto a PET film. A dilute WS₂ suspension was vacuum filtered using a PVDF membrane with 20 nm pores (Millipore). Subsequently, the WS₂/PVDF film was dried at room temperature.

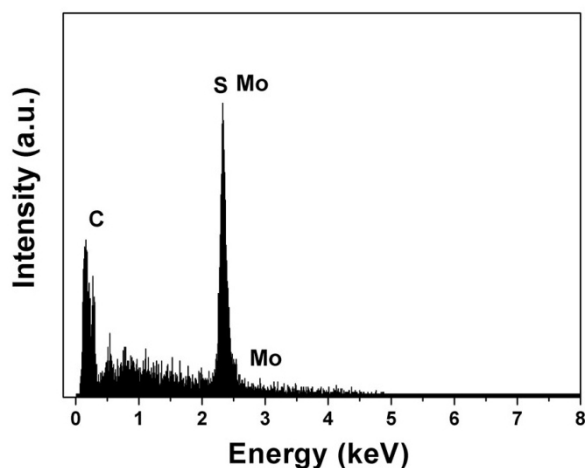


Figure S8 EDS spectra of the MoS₂ thin films on PET substrate. Elemental analysis of exfoliated MoS₂ sample using the EDS spectrum of showing the presence of Mo and S.

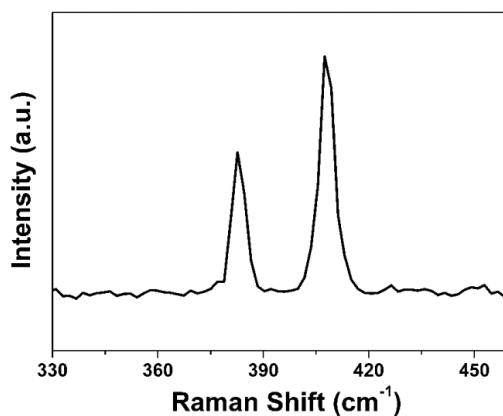


Figure S9 Raman spectra of the MoS₂ thin films on PET substrate.

S6. Comparison between our method and the previous liquid-phase exfoliation methods

Table S1. Comparison between our method and the previous liquid-phase exfoliation methods

ref	method	Cost and time	Single-layer yield	flake size	toxicity	recycle	crystallinity
<i>Nano Lett.</i> 2011, 11 , 5111; <i>ACS Nano</i> 2012, 6 , 7311	Ion intercalation using n-butyllithium, air-free; elevated reaction temperature (100 °C)	Expensive reagent; time consuming	low	Small,	High	hard	poor
<i>Nat. Commun.</i> 2014, 5 , 2995	Ion intercalation using sodium naphthalenide; air-free; pre-treated with N ₂ H ₄	two steps, time consuming	high	large	using toxic N ₂ H ₄	Naphthalenide is hard to recycle	high
<i>Science</i> 2011, 331, 568; <i>Adv. Mater.</i> 2011, 23, 3944	Ultrasonication in solvents or aqueous surfactant solutions	Expensive organic solvents, long time sonication	low	small	low	Organic solvents is hard to recycle	high
<i>Angew. Chem., Int. Ed.</i> 2011, 50, 10839	Mixed-solvent strategy for liquid exfoliation	Low cost, long time sonication	low	small	low	easy	high
Our method	Directly exfoliation using liquid alkali metal alloys at room temperature; without pre-treated	Low cost, one step	high	large	low	easy	high