Electronic Supplementary Information (ESI) for

Grain boundary engineering in atomically-thin nanosheets achieving bright white light emission

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S1. Small-angle XRD patterns and the FE-SEM image of the $Zn_{1-x}Cd_xS$(ha) hybrid precursors.

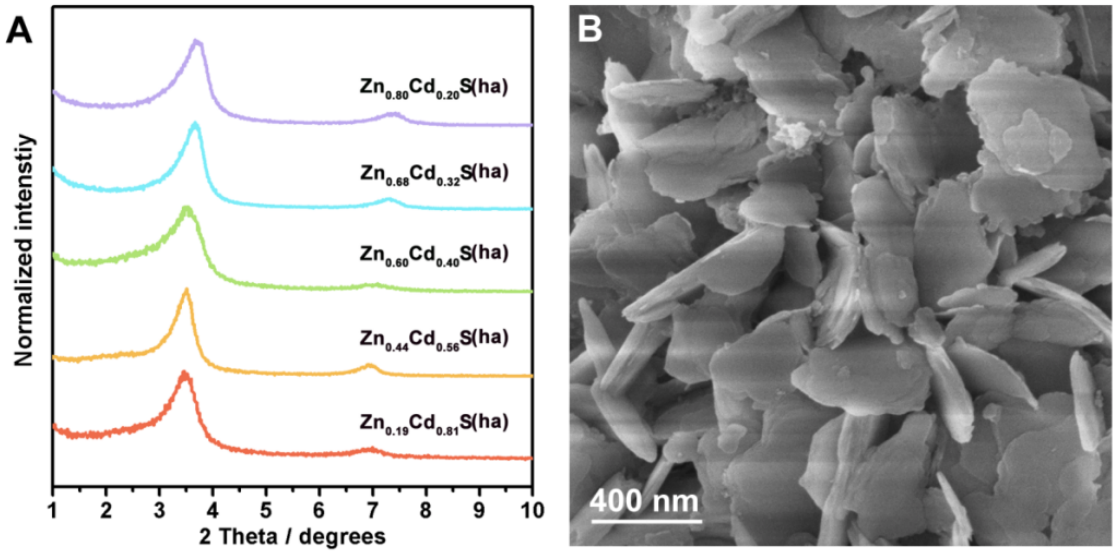

Fig. S1 (A) Small-angle XRD patterns and (B) FE-SEM image of the $Zn_{1-x}Cd_xS$(ha) hybrid precursors, revealing the typical layered structure. The layered structure of the precursors plays a crucial role in fabricating the non-layered atomically-thin nanosheets.

S2. Small-angle XRD patterns and the FE-SEM image of the grain boundary-rich atomically-thin $Zn_{1-x}Cd_xS$ nanosheets.

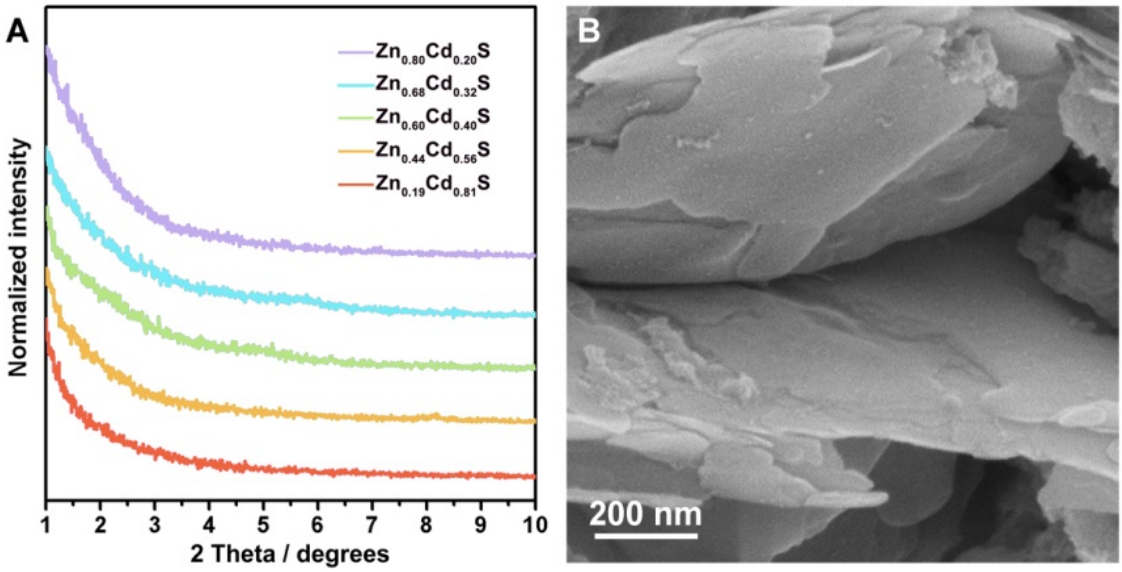

Fig. S2 (A) Small-angle XRD patterns and (B) FE-SEM image of the grain boundary-rich atomically-thin $Zn_{1-x}Cd_xS$ nanosheets, revealing the complete removal of ha molecules and the ultrathin nanosheet morphology.

S3. Photos of the dispersions of the atomically-thin $Zn_{1-x}Cd_xS$ nanosheets with grain boundary engineering in various solvents.

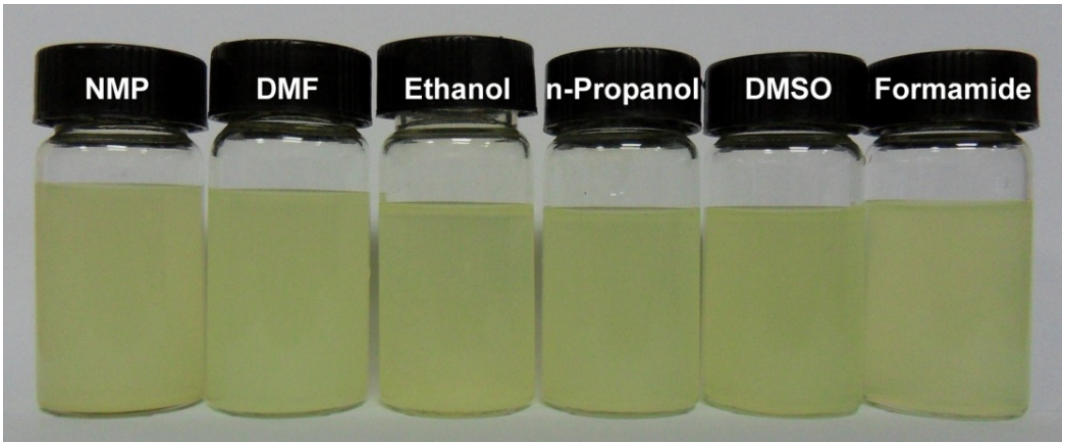

Fig. S3 Liquid dispersions of the atomically-thin $Zn_{1-x}Cd_xS$ nanosheets with grain boundary engineeing in various solvents. Solvents from left to right: N-methyl-pyrrolidone (NMP), N-dimethylformamide (DMF), ethanol, n-propanol, dimethyl sulfoxide (DMSO) and formamide.

S4. Photos of the transferable thin-films on arbitrary substrates.

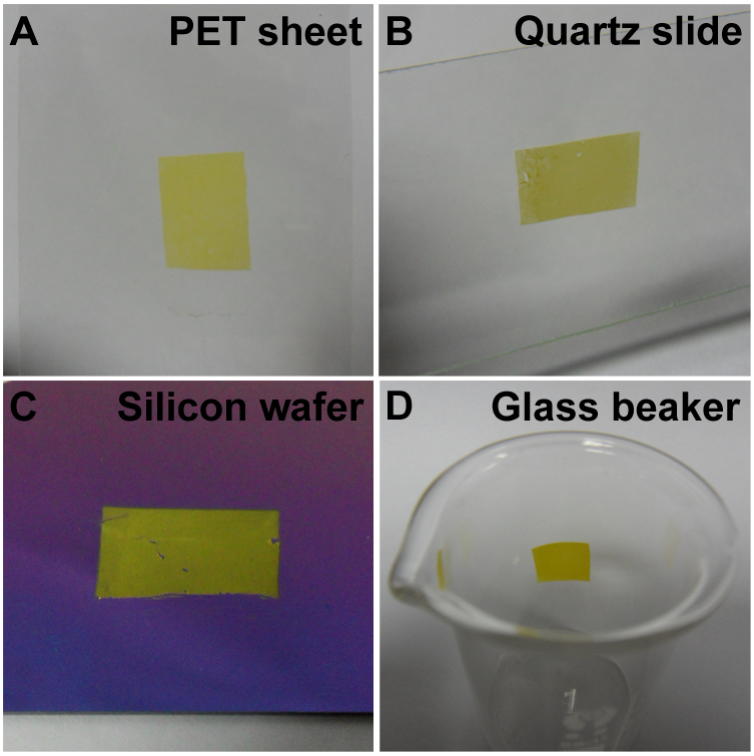

Fig. S4 Photos of the transferable thin-films on arbitrary substrates such as PET sheet, quartz slide, silicon wafer as well as the inner surface of a glass beaker. The excellent transferability and flexibility make the as-synthesized atomically-thin nanosheets promising candidates for practical utilization.

S5. Detailed calculation of the bandgap.

In order to obtain the bandgap, the following relational expression is applied:[1-3]

$(\alpha h\nu)^{1/n} = A(h\nu - E_g)$

where h is the Planck's constant, ν is the frequency of vibration, α is the absorption coefficient, E_g is the bandgap and A is the proportional constant. The value of the exponent n denotes the nature of the sample transition. In this work, the ternary materials with direct bandgap possess the exponent n=1/2. Therefore, Tauc plots can be obtained as shown in Fig. 2B in the main text, where the corresponding bandgap can be directly estimated (Table S1).

Table S1. List of the calculated bandgap of the atomically-thin $Zn_{1-x}Cd_xS$ nanosheets with different composition.

Materials	Calculated bandgap (eV)
$Zn_{0.80}Cd_{0.20}S$	3.05
$Zn_{0.68}Cd_{0.32}S$	2.87
$Zn_{0.60}Cd_{0.40}S$	2.66
$Zn_{0.44}Cd_{0.56}S$	2.59
$Zn_{0.19}Cd_{0.81}S$	2.52

References

[1] Tauc, J.; Grigorovici, R.; Vancu, A. *Phys. Status Solidi* **1966**, *15*, 627.

[2] Tauc, J. *Optical Properties of Solids*, (Eds: F. Abeles), North-Holland, **1972**.

[3] Davis, E. A.; Mott, N. F. *Philos. Mag.* **1970**, *22*, 903.