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$_x$S(ha) hybrid precursors.

**Fig. S1** (A) Small-angle XRD patterns and (B) FE-SEM image of the Zn$_{1-x}$Cd$_x$S(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$_x$S nanosheets.

**Fig. S2** (A) Small-angle XRD patterns and (B) FE-SEM image of the grain boundary-rich atomically-thin Zn$_{1-x}$Cd$_x$S 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$_x$S nanosheets with grain boundary engineering in various solvents.

Fig. S3 Liquid dispersions of the atomically-thin Zn$_{1-x}$Cd$_x$S nanosheets with grain boundary engineering 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, \( \nu \) is the frequency of vibration, \( \alpha \) 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 \( \text{Zn}_{1-x}\text{Cd}_x\text{S} \) nanosheets with different composition.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Calculated bandgap (eV)</th>
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<tbody>
<tr>
<td>( \text{Zn}<em>{0.80}\text{Cd}</em>{0.20}\text{S} )</td>
<td>3.05</td>
</tr>
<tr>
<td>( \text{Zn}<em>{0.68}\text{Cd}</em>{0.32}\text{S} )</td>
<td>2.87</td>
</tr>
<tr>
<td>( \text{Zn}<em>{0.60}\text{Cd}</em>{0.40}\text{S} )</td>
<td>2.66</td>
</tr>
<tr>
<td>( \text{Zn}<em>{0.44}\text{Cd}</em>{0.56}\text{S} )</td>
<td>2.59</td>
</tr>
<tr>
<td>( \text{Zn}<em>{0.19}\text{Cd}</em>{0.81}\text{S} )</td>
<td>2.52</td>
</tr>
</tbody>
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**References**