

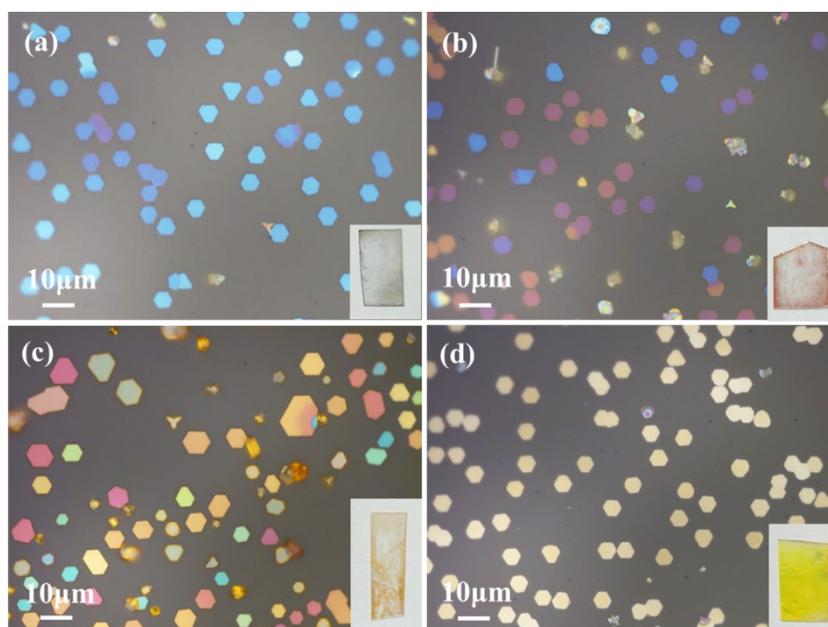
## Supporting Information

### Van der Waals Epitaxial Two-Dimensional $\text{CdS}_x\text{Se}_{(1-x)}$ Semiconductor Alloys with Tunable-Composition and Application to Flexible Optoelectronics

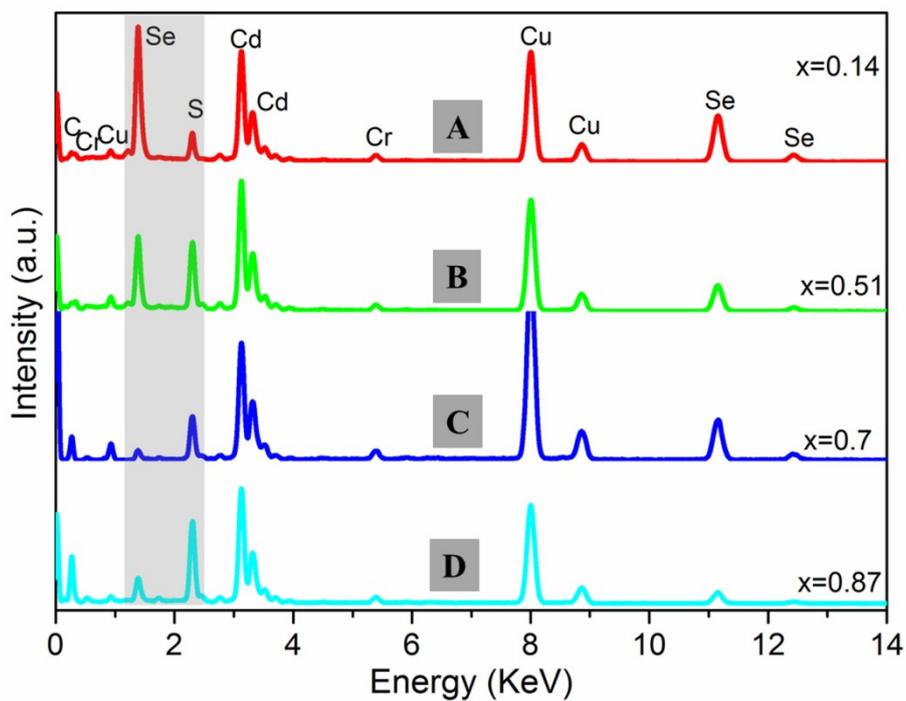
Jing Xia, Yun-Xuan Zhao, Lei Wang, Xuan-Ze Li, Yi-Yi Gu, Hua-Qiu Cheng and Xiang-Min Meng\*

Key Laboratory of Photochemical Conversion and Optoelectronic Materials, Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing, 100190, P. R. China

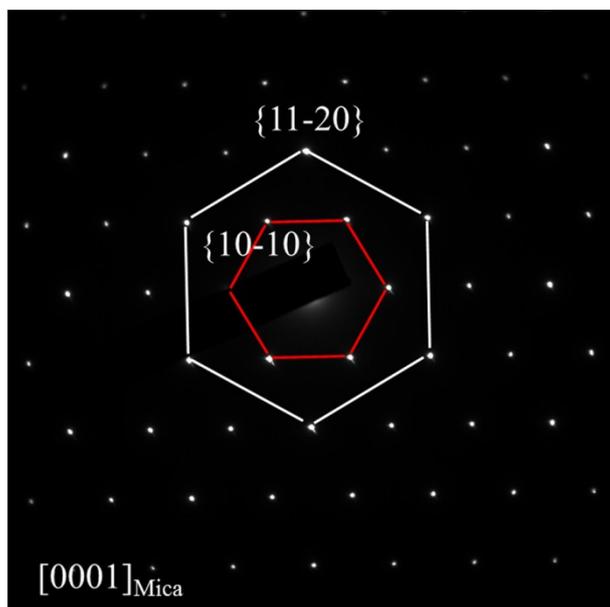
E-mail of corresponding author: [mengxiangmin@mail.ipc.ac.cn](mailto:mengxiangmin@mail.ipc.ac.cn)



**Fig. S1** (a-d) Optical images of the 2D  $\text{CdS}_x\text{Se}_{(1-x)}$  ( $x = 0.14, 0.51, 0.7$  and  $0.87$ ) samples, respectively. Insets show the photograph of the 2D  $\text{CdS}_x\text{Se}_{(1-x)}$  alloy flakes on mica substrates, respectively.



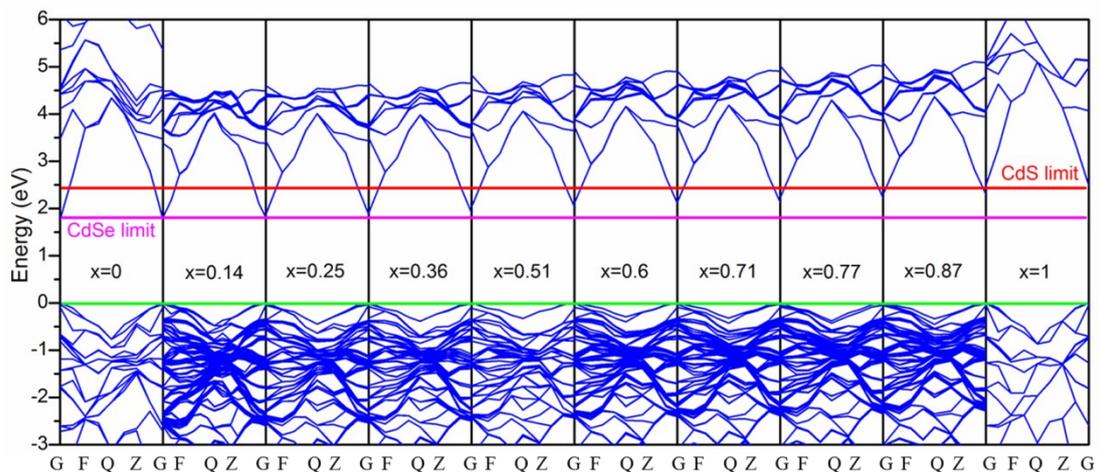
**Fig. S2** EDS spectra of the  $\text{CdS}_x\text{Se}_{(1-x)}$  alloys with different chemical compositions. Curves A-D come from the samples in Fig. S1a-d, respectively.



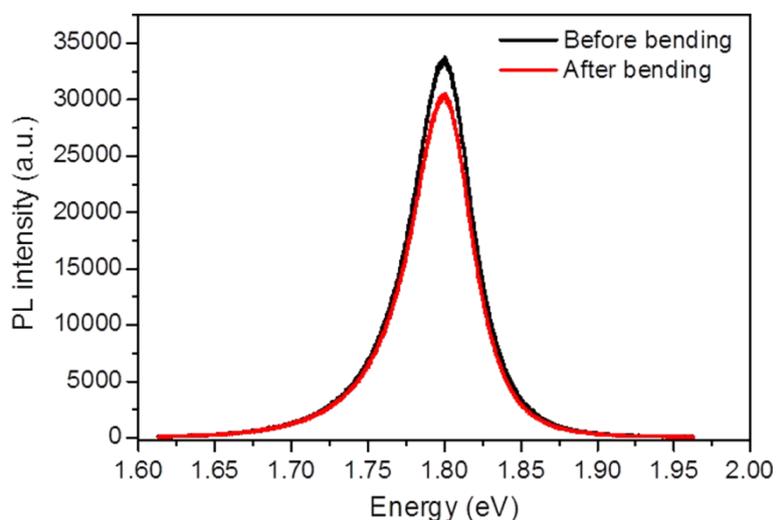
**Fig. S3** SAED pattern of the fluorophlogopite mica sheet.

Overlayer/Substrate	Lattice Parameter a	Matching Relationship	Lattice Mismatch $f$
CdS <sub>0.14</sub> Se <sub>0.86</sub> /mica	0.422 nm/0.53nm	(0001)[11-20] <sub>CdS<sub>x</sub>Se<sub>(1-x)</sub></sub>    (0001)[11-20] <sub>mica</sub>	20.3%
		(0001)[1-100] <sub>CdS<sub>x</sub>Se<sub>(1-x)</sub></sub>    (0001)[1-100] <sub>mica</sub>	20.3%
		(0001)[11-20] <sub>CdS<sub>x</sub>Se<sub>(1-x)</sub></sub>    (0001)[2-200] <sub>mica</sub>	30.9%
		(0001)[2-200] <sub>CdS<sub>x</sub>Se<sub>(1-x)</sub></sub>    (0001)[11-20] <sub>mica</sub>	8.3%
CdS <sub>0.51</sub> Se <sub>0.49</sub> /mica	0.418nm/0.53nm	(0001)[11-20] <sub>CdS<sub>x</sub>Se<sub>(1-x)</sub></sub>    (0001)[11-20] <sub>mica</sub>	21.1%
		(0001)[1-100] <sub>CdS<sub>x</sub>Se<sub>(1-x)</sub></sub>    (0001)[1-100] <sub>mica</sub>	21.1%
		(0001)[11-20] <sub>CdS<sub>x</sub>Se<sub>(1-x)</sub></sub>    (0001)[1-100] <sub>mica</sub>	31.7%
		(0001)[1-100] <sub>CdS<sub>x</sub>Se<sub>(1-x)</sub></sub>    (0001)[11-20] <sub>mica</sub>	8.7%
CdS <sub>0.7</sub> Se <sub>0.3</sub> /mica	0.417nm/0.53nm	(0001)[11-20] <sub>CdS<sub>x</sub>Se<sub>(1-x)</sub></sub>    (0001)[11-20] <sub>mica</sub>	21.3%
		(0001)[1-100] <sub>CdS<sub>x</sub>Se<sub>(1-x)</sub></sub>    (0001)[1-100] <sub>mica</sub>	21.3%
		(0001)[11-20] <sub>CdS<sub>x</sub>Se<sub>(1-x)</sub></sub>    (0001)[1-100] <sub>mica</sub>	32.1%
		(0001)[1-100] <sub>CdS<sub>x</sub>Se<sub>(1-x)</sub></sub>    (0001)[11-20] <sub>mica</sub>	9.2%
CdS <sub>0.87</sub> Se <sub>0.13</sub> /mica	0.415nm/0.53nm	(0001)[11-20] <sub>CdS<sub>x</sub>Se<sub>(1-x)</sub></sub>    (0001)[11-20] <sub>mica</sub>	21.7%
		(0001)[1-100] <sub>CdS<sub>x</sub>Se<sub>(1-x)</sub></sub>    (0001)[1-100] <sub>mica</sub>	21.7%
		(0001)[11-20] <sub>CdS<sub>x</sub>Se<sub>(1-x)</sub></sub>    (0001)[1-100] <sub>mica</sub>	32.2%
		(0001)[1-100] <sub>CdS<sub>x</sub>Se<sub>(1-x)</sub></sub>    (0001)[11-20] <sub>mica</sub>	9.6%

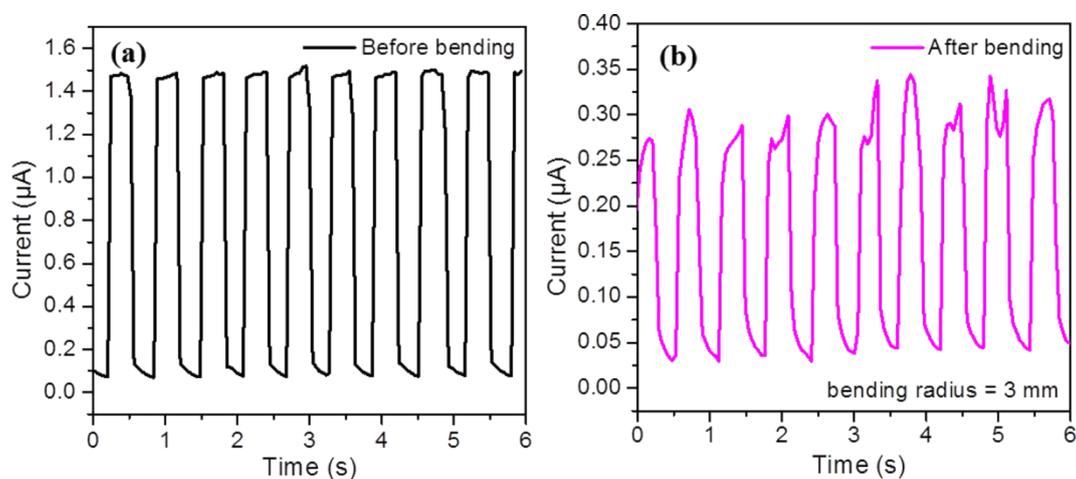
**Table S1** Calculated lattice mismatches of the 2D CdS<sub>x</sub>Se<sub>(1-x)</sub> alloys on the fluorophlogopite mica sheet.



**Fig. S4** Calculated band structure of the 2D CdS<sub>x</sub>Se<sub>(1-x)</sub> alloy samples with varying composition x.



**Fig. S5** Room-temperature PL spectra of the  $\text{CdS}_{0.14}\text{Se}_{0.86}$  flake from the flexible device in Fig. 6 before and after bending for 50 times.



**Fig. S6** (a) and (b) Time-dependent photoresponse of a  $\text{CdS}_{0.51}\text{Se}_{0.49}$  flake flexible photodetector before and after bending for 50 times. Inset shows the photograph of the setup for the bending measurements. The photoresponse test was performed under 450 nm laser illumination with the light intensity of  $4.6 \text{ mW/cm}^2$  and bias of 5 V.