Supporting Information for

A Universal Atomic Substitution Conversion Strategy Towards

Synthesis of Large-Size Ultrathin Nonlayered Two-Dimensional

Materials

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Supplementary Figures



Fig. S1 Dark-field OM image (**a**) and Raman spectrum (**b**) of large-size ultrathin CdS flake in Fig. 3c

Nano-Micro Letters



Fig. S2 Typical bright-field and dark-field OM images of large-area CdI_2 flakes with various thickness before (a) and after (b, c) conversion into CdS flakes



Fig. S3 Typical OM image (**a**) and corresponding AFM image (**b**), and Raman intensity mapping of converted CdS flakes with different thickness (**c**)



Fig. S4 a Raman spectra of CdBr₂ flakes grown on SiO₂/Si substrate and corresponding converted CdS flakes. **b** XRD pattern of CdBr₂ flakes grown on SiO₂/Si substrate and corresponding converted CdS flakes. **c** Raman spectra of CdI₂ flakes grown on SiO₂/Si substrate and corresponding converted CdSe flakes. **d** XRD pattern of CdI₂ flakes grown on SiO₂/Si substrate and corresponding converted CdSe flakes. **d** XRD pattern of CdI₂ flakes grown on SiO₂/Si substrate and corresponding converted CdSe flakes. **d** XRD pattern of CdI₂ flakes grown on SiO₂/Si substrate and corresponding converted CdSe flakes.



Fig. S5 OM image and corresponding AFM height profile of the converted CdS flakebased photodetector via the transfer electrode method