Supporting Information:

Manipulation of Valley pseudospin Polarization by Selective Spin Injection in Chiral Two-Dimensional Perovskite/Monolayer Transition Metal Dichalcogenide Heterostructures

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Figure S1. Fluorescence image (a) and Raman spectrum (b) of a monolayer MoS$_2$ in Figure 1b. The excitation source is a 532 nm laser with the power of 178 μW. The scale bar is 10 μm.
Figure S2. Lorentz fittings of PL spectra. (a) PL spectrum of a monolayer MoS$_2$ fitted with four Lorentz peaks, which are neutral A exciton (X), negatively charged A trion (X$^-$), B exciton and defect-induced emission (D). (b) PL spectra of monolayer MoS$_2$ and MoS$_2$ heterostructure corresponding to Figure 1c. To be noticed, all spectra are fitted with four Lorentz peaks but only show the X and X$^-$ peaks. The X$^-$ weight can be determined by $\gamma = \frac{I_{X^-}}{I_{X^-} + I_X}$, where $\gamma$ is the X$^-$ weight, $I_{X^-}$ is the area of X$^-$ peak and $I_X$ is the area of X peak.
Figure S3. Optical setup of the polarization-resolved PL measurement. The excitation laser first passes through a beam splitter to excite the sample and then the emission signal was detected by a set of quarter-wave plate, half-wave plate and another polarizer.
Figure S4. Polarization-resolved PL spectra of bare (R-MBA)$_2$PbI$_4$ and (R-MBA)$_2$PbI$_4$ in (R-MBA)$_2$PbI$_4$/MoS$_2$ (a), bare (S-MBA)$_2$PbI$_4$ and (S-MBA)$_2$PbI$_4$ in (S-MBA)$_2$PbI$_4$/MoS$_2$ (b) excited by a linearly polarized 473 nm laser at 78 K. The spectra are taken from other two representative devices different from the ones used in Figure 1 in the main text.
Figure S5. Polarization-resolved PL spectra of (R-MBA)$_2$PbI$_4$ excited by linearly polarized 473 nm, 532 nm and 633 nm lasers at 78 K.
Figure S6. Power-dependent polarization-resolved PL (a) and the calculated absolute DOP (b) of a (S-MBA)$_2$PbI$_4$/MoS$_2$ heterostructure at 78 K excited by a linearly polarized 532 nm laser with the power of 6.43 mW.
Figure S7. Temperature-dependent PL studies of a (R-MBA)$_2$PbI$_4$/MoS$_2$ heterostructure.

Temperature-dependent spectra of monolayer MoS$_2$ (a) and the (R-MBA)$_2$PbI$_4$/MoS$_2$ heterostructure (Het.) (b) excited by a linearly polarized 532 nm laser with the power of 6.43 mW. (c) The peak position shifts of A exciton in MoS$_2$ and the (R-MBA)$_2$PbI$_4$/MoS$_2$ heterostructure versus temperature. The black and red dashed lines are fitted by the Varshni equation,$^{1,2}$ which describes the band gap evolution with temperature.
Figure S8. Polarization-resolved PL spectra of (R-MBA)$_2$PbI$_4$ (a), (S-MBA)$_2$PbI$_4$ (b) and (rac-MBA)$_2$PbI$_4$ (c) microplates used in Figure 4a excited by a linearly polarized 473 nm laser with the power of 0.02 μW at 78 K.
Figure S9. Band alignment of monolayer MoS$_2$, monolayer WSe$_2$ and chiral 2D perovskite as well as the charge transfer and recombination process in the (R-MBA)$_2$PbI$_4$/WSe$_2$ heterostructure.
Figure S10. PL spectra of an edge excited (R-MBA)$_2$PbI$_4$/WSe$_2$ heterostructure at 78 K. (a) PL spectrum of (R-MBA)$_2$PbI$_4$ in the upper panel and its CPL emission spectra in the lower panel excited by a linearly polarized 473 nm laser with the power of 0.02 μW. (b) Polarization-resolved PL of the constituent monolayer WSe$_2$ (upper panel) and the (R-MBA)$_2$PbI/WSe$_2$ heterostructure (lower panel) excited by a linearly polarized 633 nm laser with the power of 485 μW. Inset: schematic illustration of the edge excitation with the excitation region highlighted by the red box.
References
