Controllable Valley Magnetic Response in Phase Transformed Tungsten Diselenide

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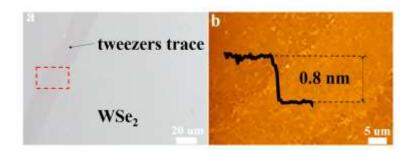


Fig. S1 (a) Optical topography of the CVD grown large-scale WSe₂ monolayer on SiO₂/Si surface, where the tweezers trace shows the distinction between WSe₂ film and the substrate. (b) AFM image taken in the area denoted by the red dashed box in (a).

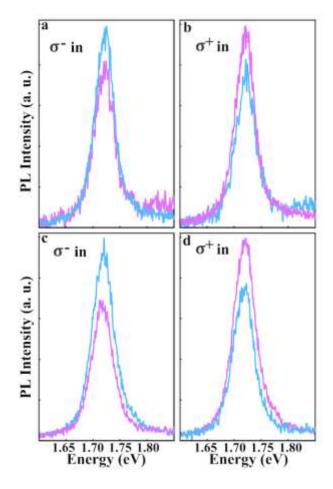


Fig. S2 Circularly-polarized PL spectra of (a,b) H/T-WSe₂-12.2% under and (c,d) H/T-WSe₂-19.9%. All the spectra are collected under 10K without magnetic field. The blue and purple lines indicate co- and cross-polarized configurations, respectively.

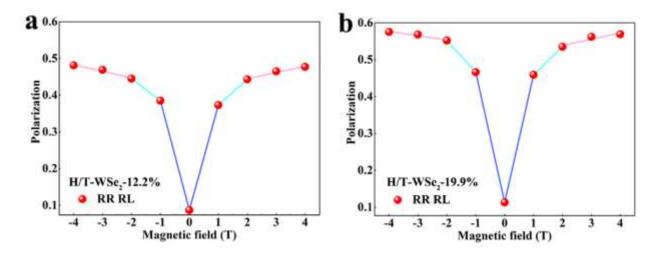


Fig. S3 Magnetic-field dependent polarization of (a) H/T-WSe₂-12.2% and (b) H/T-WSe₂-19.9% under σ^+ excitation with σ^+ and σ^- detection.

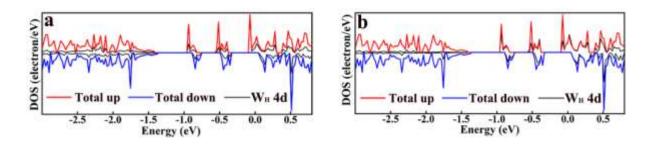


Fig. S4 Spin-projected density of states of W-4d orbitals in the (a) H phase and (b) T phase of H/T-WSe₂.

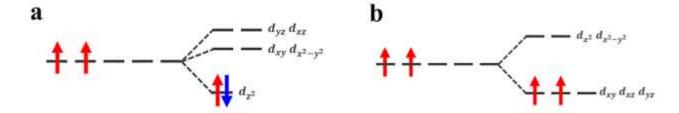


Fig. S5 The occupation of electrons in W-4d orbitals under the crystal fields of (a) H phase and (b) T phase WSe₂.

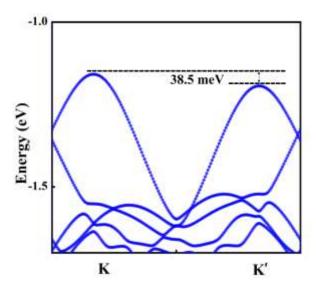


Fig. S6 Spin-projected band structures of monolayer H/T-WSe₂-19.9%, which shows a valley splitting of 38.5 meV.

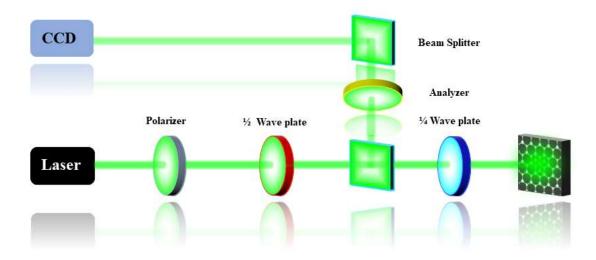


Fig. S7 (a) Optical valley-polarized emission of H/T-WSe₂ produced by the polarization-resolved PL microscope system.