Supplementary Information

2D ternary vanadium phosphorous chalcogenide with strong in-plane

optical anisotropy

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Fig. S1 (a) The unit cell structure of triclinic $V_2P_4S_{13}$. (b) Lattice parameters of $V_2P_4S_{13}$.



Fig. S2 Top view of distinctive porous 2D $V_2P_4S_{13}$ with anisotropic distribution of angstrom-scale pores indicated by different fill colors.



Fig. S3 Scanning electron microscope image of single crystal $V_2P_4S_{13}$.



Fig. S4 (a) Energy dispersive spectrum of $V_2P_4S_{13}$ nanosheet on Cu grid. (b) Quantitative analysis results of energy dispersive spectrum of $V_2P_4S_{13}$ nanosheet.



Fig. S5 Optical microscope image of $V_2P_4S_{13}$ flake for angle-resolved polarized Raman spectroscopy measurements.

Table S1 Polar plots of selected Raman modes under parallel and verticalpolarization configurations.





Fig. S6 Optical microscope image of $V_2P_4S_{13}$ flake under transmittance mode for polarization-resolved absorption spectroscopy measurements.