

Supplementary Information for

Tunable ferrovalley semiconductors of Janus VSeCl and VSeBr

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Table S1 The calculated lattice constant (a), magnetic moments of V (m_V), energy difference between AFM and FM states per unit cell (ΔE), magnetic exchange interaction parameter (J), magnetic anisotropy energy (MAE), band gap (E_g), Curie Temperature (T_C), and valley polarization at the valence band ($\Delta E_V^{K\pm}$) and conduction band ($\Delta E_C^{K\pm}$) for ML VSeCl and VSeBr.

	a (Å)	m_V (μ_B)	ΔE (meV)	J (S=1)	MAE (meV)	E_g (eV)	T_C (K)	$\Delta E_V^{K\pm}$ (meV)	$\Delta E_C^{K\pm}$ (meV)
VSeCl	3.59	1.96	23.63	5.91	0.171	0.41	76	68.3	3.4
VSeBr	3.69	1.97	33.66	8.42	0.148	0.49	106	65.1	1.8

Table S2 The calculated lattice constant (a), energy difference between AFM and FM states per unit cell (ΔE), magnetic exchange interaction parameter (J), magnetic anisotropy energy (MAE), band gap within SOC (Gap-SOC), and valley polarization values at the valence band ($\Delta E_V^{K\pm}$) and conduction band ($\Delta E_C^{K\pm}$) when different U_{eff} values are considered.

U_{eff} (eV)	a (Å)	ΔE (meV)	J (S=1)	MAE (meV)	Gap-SOC (eV)	$\Delta E_V^{K\pm}$ (meV)	$\Delta E_C^{K\pm}$ (meV)
0	3.50	18.08	4.52	-0.092	/	/	/
1.16	3.59	23.63	5.91	0.171	0.37	68.3	3.4
2	3.62	23.75	5.94	0.158	0.92	73.3	2.6
3	3.65	24.03	6.01	0.137	1.38	80.0	2.0

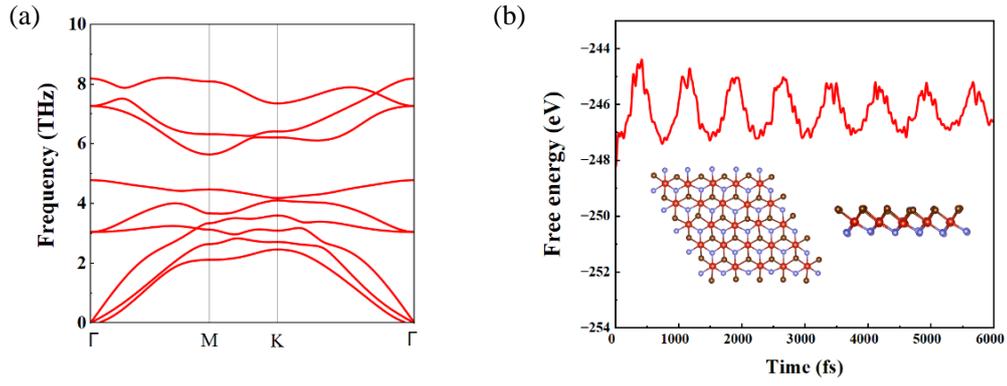


Fig. S1 (a) The phonon spectrum of the ML VSeBr. (b) Time evolution of free energy for a 4×4 supercell of ML VSeBr at 300 K in the AIMD simulation. The top and side views of the final structure are shown in the figure. The red, purple and brown spheres represent V, Se and Br.

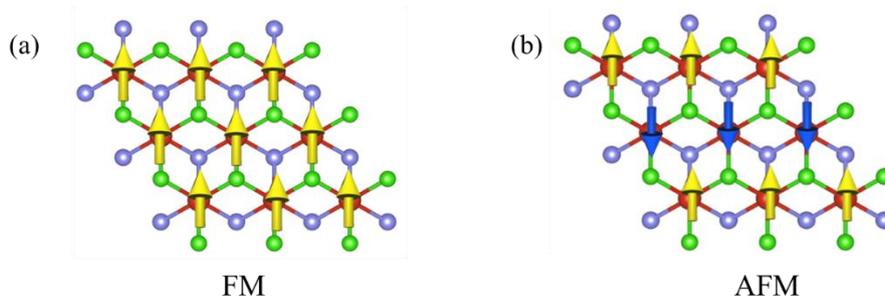


Fig. S2 (a) FM and (b) AFM states of Janus VSeCl (VSeBr). The yellow (blue) arrow shows the direction of spin-up (spin-down).

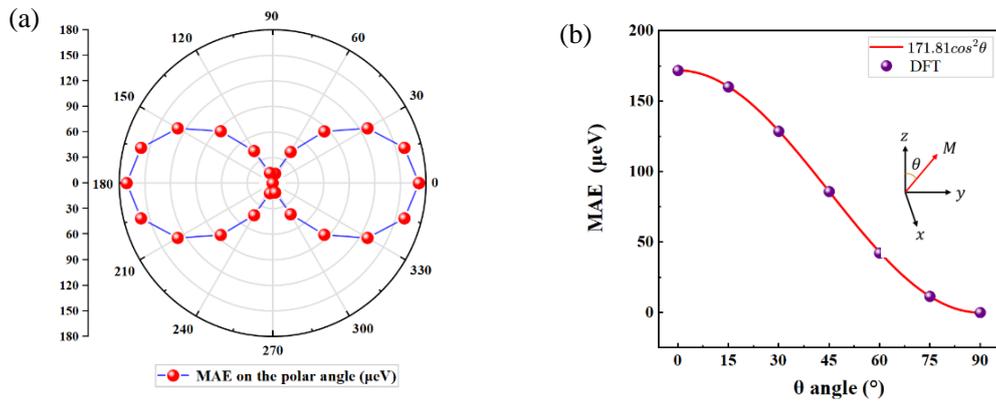


Fig. S3 (a) The variation of MAE with polar angle for Janus ML VSeCl. (b) Comparison of MAE as a function of θ between DFT and fitted equation. The inset shows the magnetization angle θ between the magnetic moment and the z-axis direction.

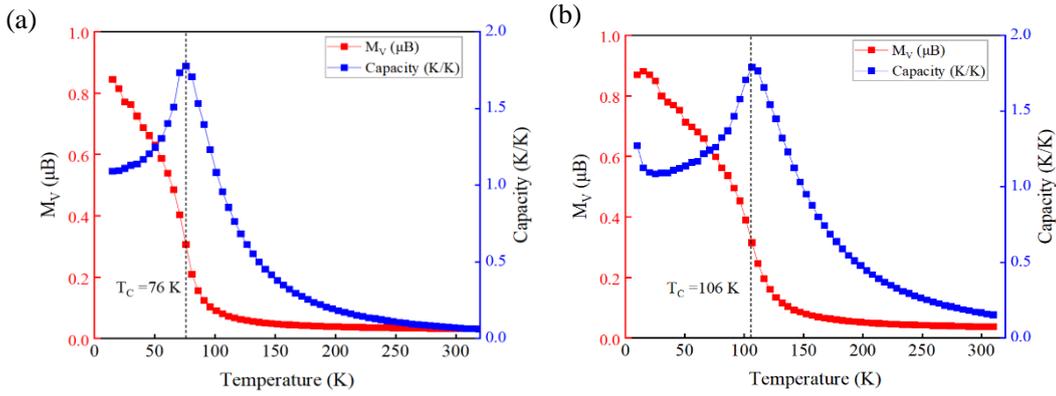


Fig. S4 The magnetic moment and specific heat capacity as a function of temperature for the ML VSeCl (a) and VSeBr (b).

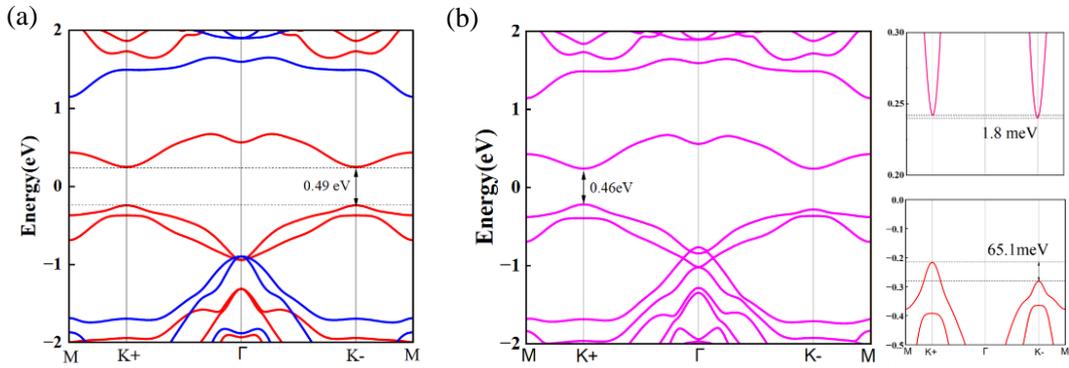


Fig. S5 The spin-dependent band structure of the ML VSeBr without (a) and with (b) SOC. Red and blue lines in (a) represent spin-up and spin-down channels, respectively. The Fermi level is set to zero.

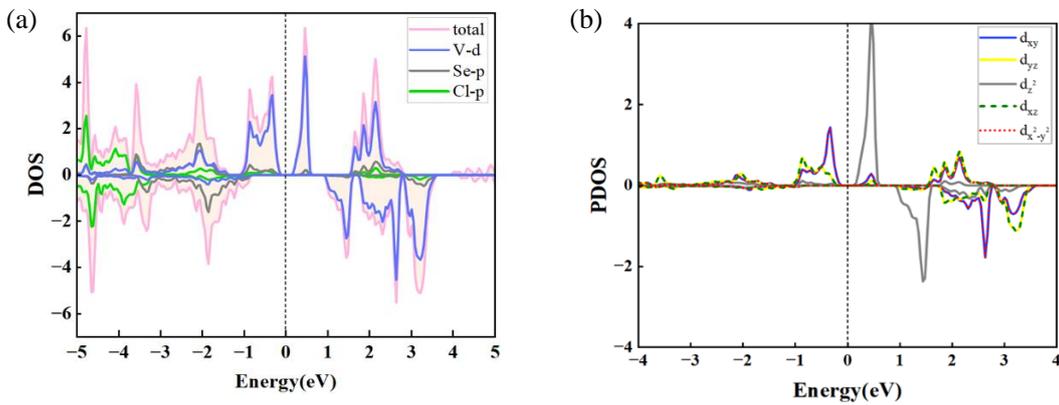


Fig. S6 (a) Density of states (DOS) of ML VSeCl. (b) Partial density of states (PDOS) for the 3d orbitals of V atom in ML VSeCl. The Fermi level is set to zero.

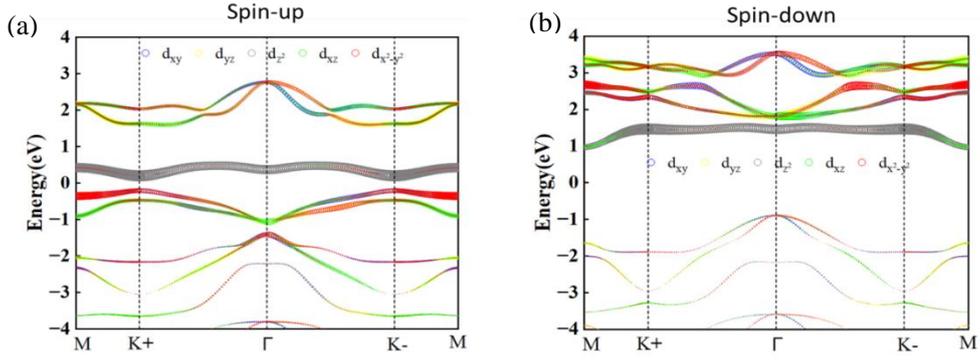


Fig. S7 Projected bands (PBAND) of the V 3d orbitals in spin-up (a) and spin-down (b) channels for ML VSeCl. The Fermi level is set to zero.

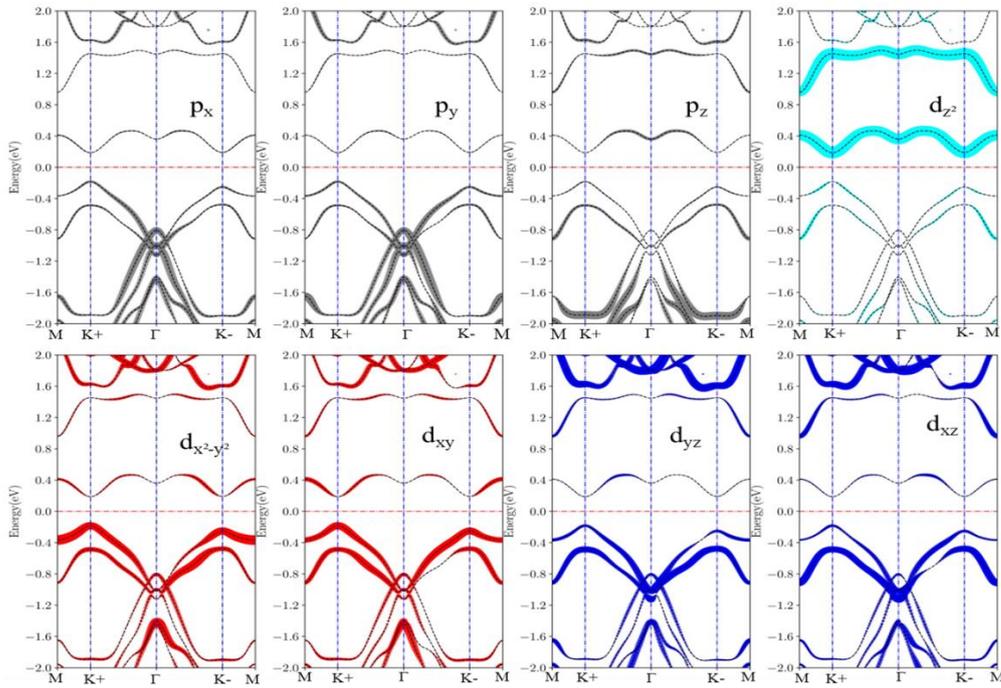


Fig. S8 Projected bands (PBAND) with SOC of p and d orbitals in ML VSeCl. The Fermi level is set to zero.

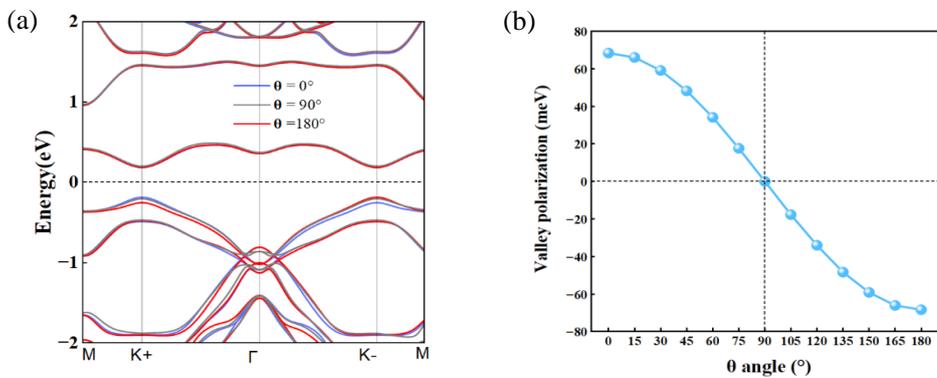


Fig. S9 (a) The band structure of ML VSeCl and (b) valley polarization at VBM controlled by tuning the polar angle.

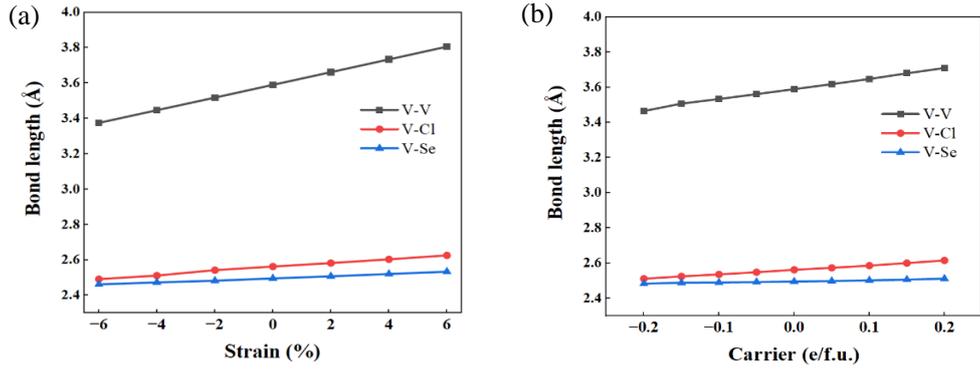


Fig. S10 Bond length of V-V, V-Cl, and V-Se as functions of strain (a) and carrier doping (b) for ML VSeCl.

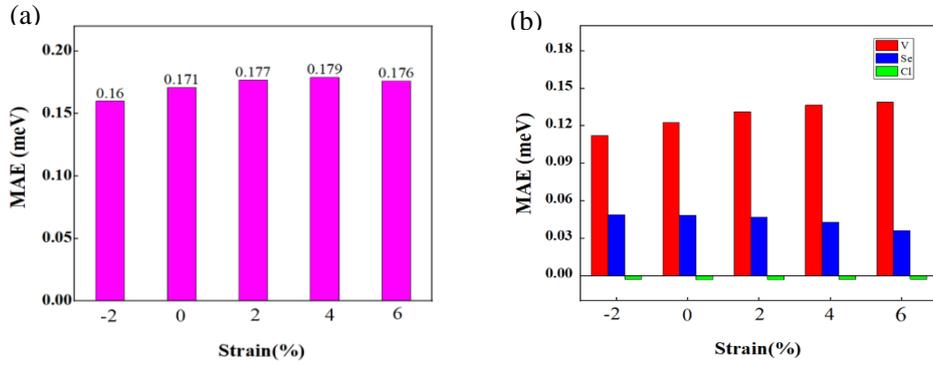


Fig. S11 The total MAE (a) and the contribution of each atom to MAE (b) as a function of strain for ML VSeCl.

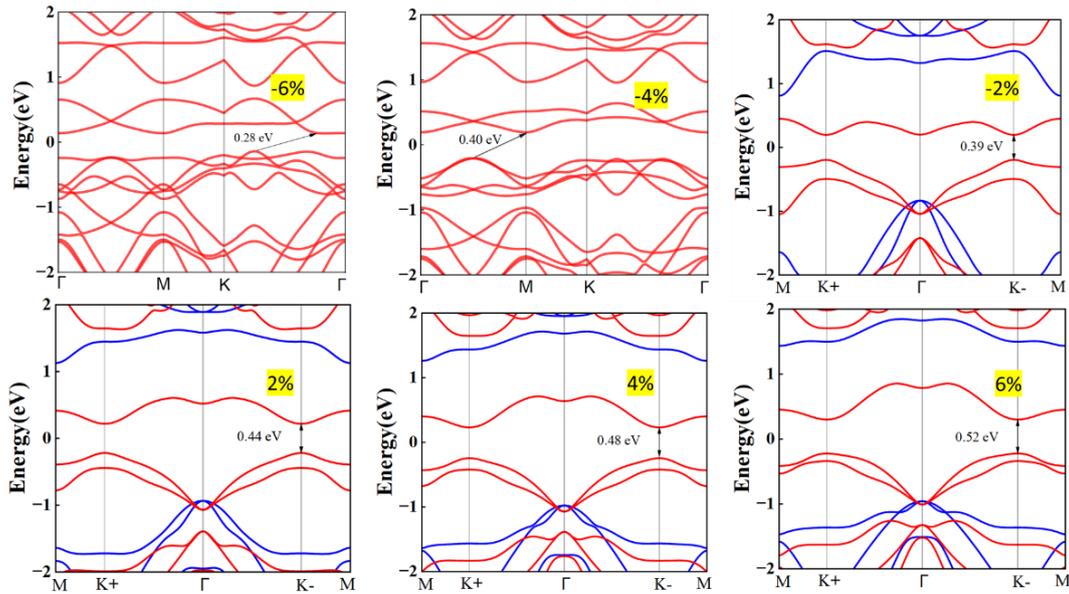


Fig. S12 Spin-polarized band structure for ML VSeCl at biaxial strains of -6% , -4% , -2% , 2% , 4% and 6% , respectively. Red and blue lines represent spin-up and spin-down channels. The Fermi level is set to zero.

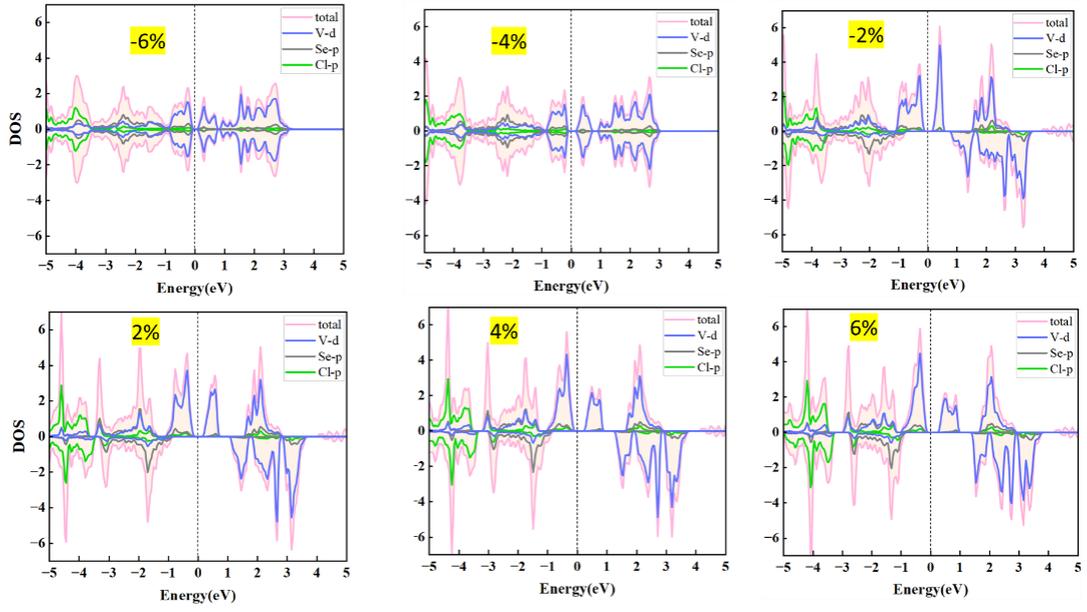


Fig. S13 Total and orbit-resolved DOS of ML VSeCl at biaxial strains of -6% , -4% , -2% , 2% , 4% and 6% , respectively.

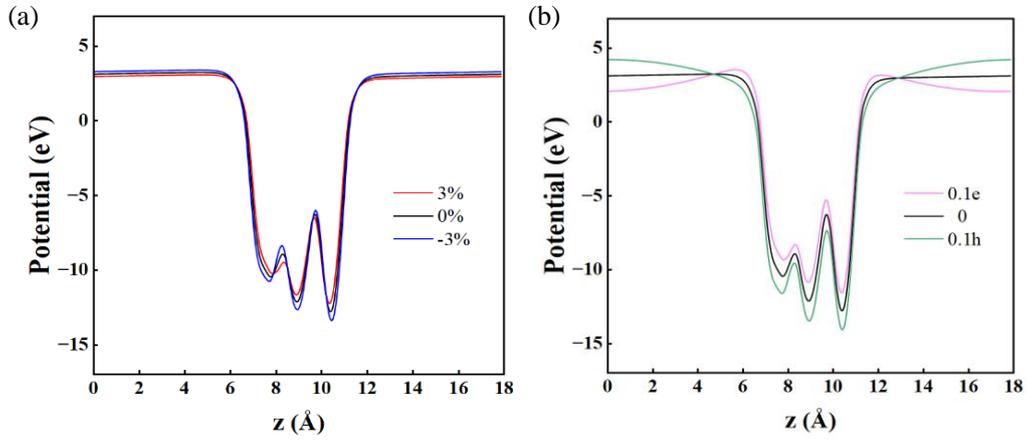


Fig. S14 (a) The planar electrostatic potential along the z -axis direction of ML VSeCl at biaxial strains of -3% , 0% and 3% . (b) The planar electrostatic potential along the z -axis direction of ML VSeCl with carrier doping of 0.1 h, 0 and 0.1 e.

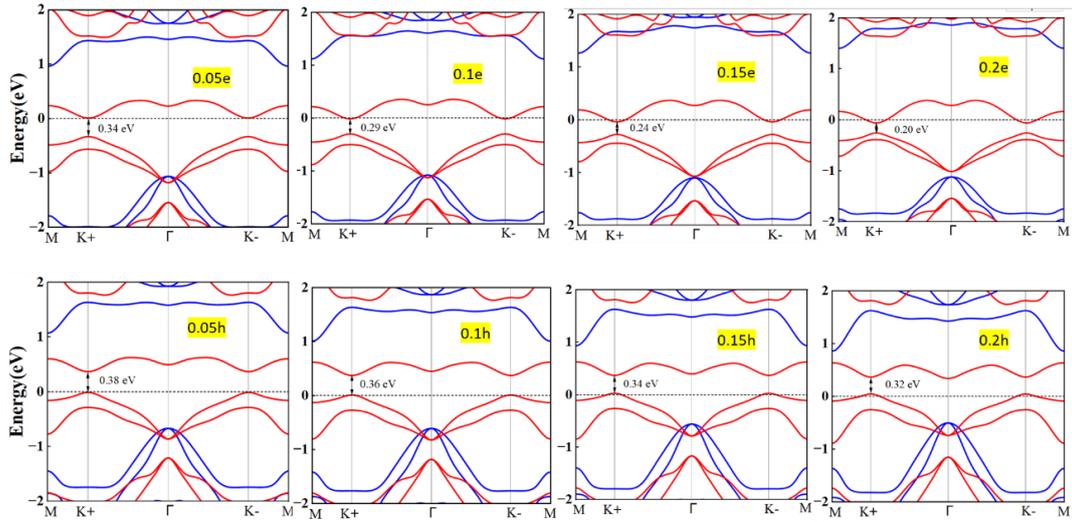


Fig. S15 Spin-polarized band structure for ML VSeCl with carrier doping of 0.05 e, 0.1 e, 0.15 e, 0.2 e, 0.05 h, 0.1 h, 0.15 h and 0.2 h, respectively. Red and blue lines represent spin-up and spin-down channels, respectively. The Fermi level is set to zero.

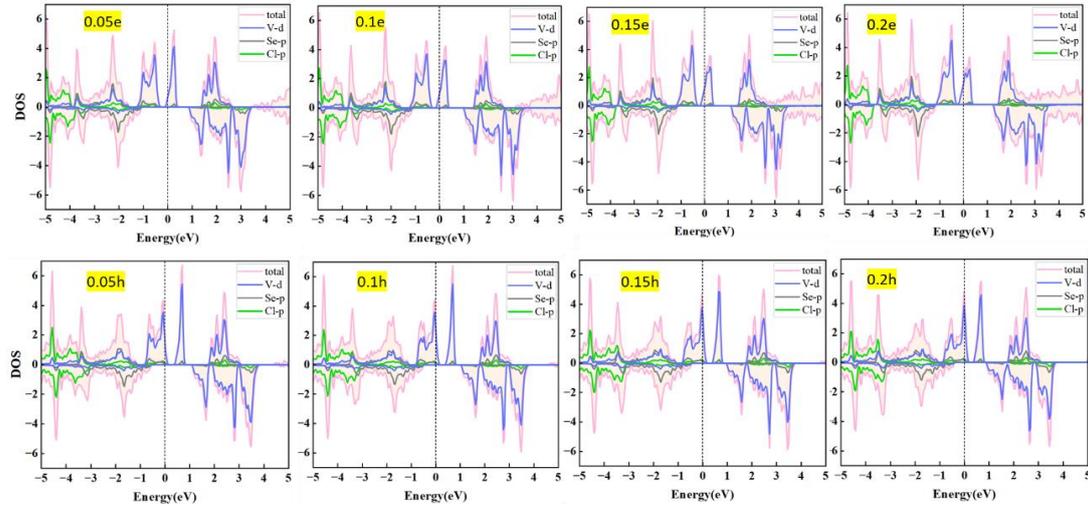


Fig. S16 The total and orbit-resolved DOS for ML VSeCl with carrier doping of 0.05 e, 0.1 e, 0.15 e, 0.2 e, 0.05 h, 0.1 h, 0.15 h and 0.2 h, respectively.

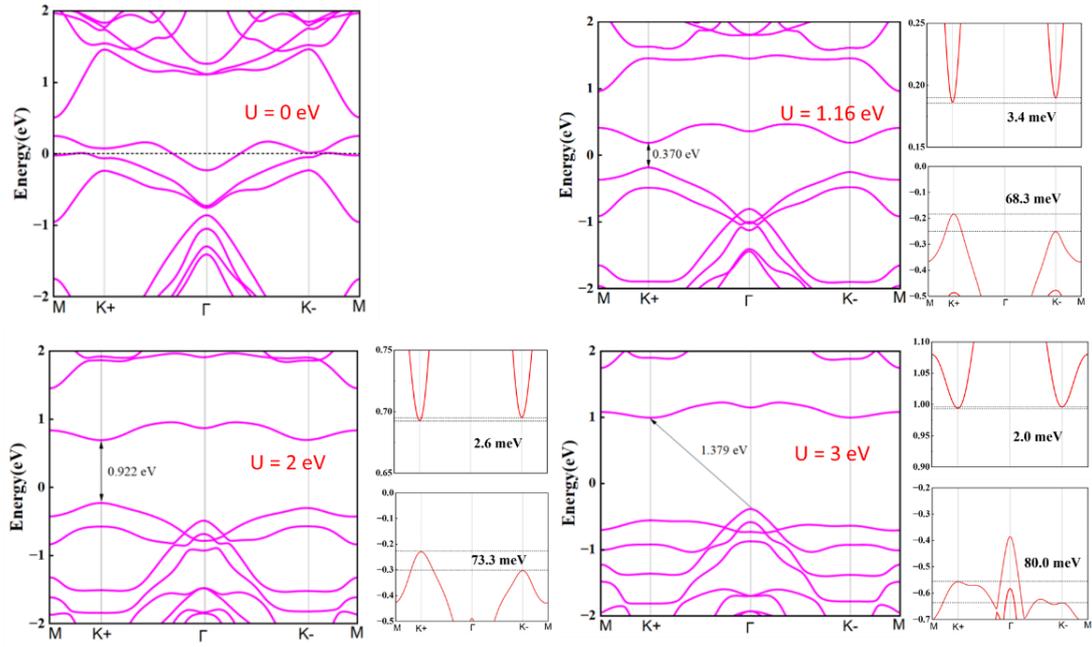


Fig. S17 Band structure within SOC for monolayer VSeCl with effective U of 0, 1.16, 2 and 3 eV, respectively. The Fermi level is set to zero.