Due to itinerant Co d-electrons, we have also considered antiferromagnetic (AFM) orderings (i.e., AFM-Néel, AFM-Stripy, AFM-Zigzag, which are shown in Fig. S1 in the supplementary material). The comparison of minimum energies, which are obtained following structural optimizations including lattice constants for each magnetic states, implies that the lowest energy configuration is FM for Co-based Janus structure. The Co atom possesses the electronic configuration of $3d^74s^2$, and $3d^7$ has unpaired electrons, which gives rise to the magnetic state of Co-based Janus structure. We should note that, favorable magnetic orientations (FM) for the considered Co-based Janus $M_4X_3Y_3$ ($M=$ Co, and $X,Y = S, Se, Te$) monolayers which are written in Table are determined by DFT calculations.
Figure S1: Magnetic moment orientations (FM, AFM-Néel, AFM-Stripy, AFM-Zigzag). Up and down arrows indicate spin-up and spin-down orientations, respectively. The table shows minimum energy configuration.