

Large perpendicular magnetic anisotropy of transition metal dimers  
driven by polarization switching of two-dimensional ferroelectric  
 $\text{In}_2\text{Se}_3$  substrate

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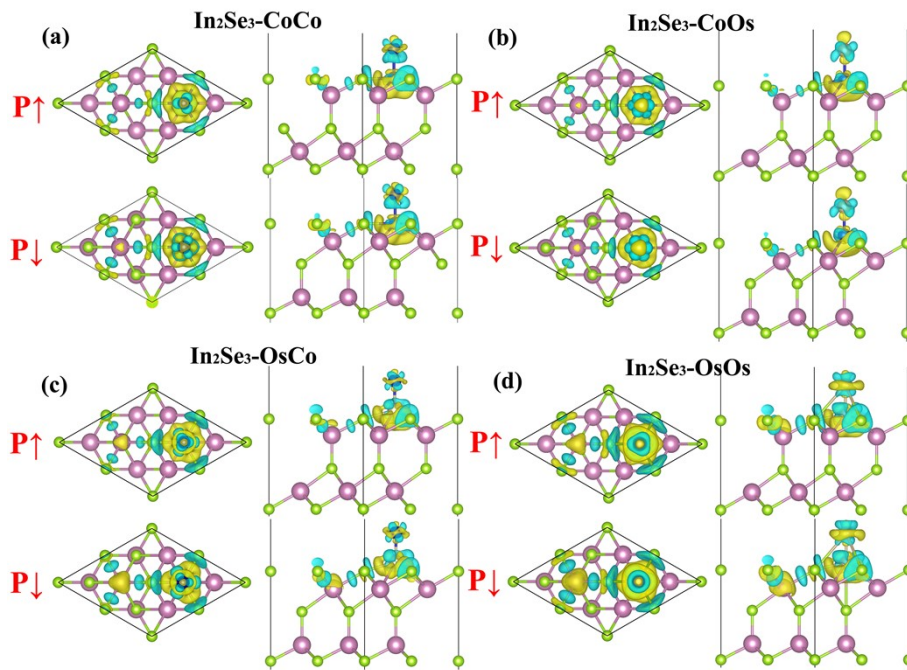


Fig. S1 Charge difference between the total charge density and the sum of charge densities of a suspended transition metal dimers and  $\text{In}_2\text{Se}_3$  for (a)  $\text{In}_2\text{Se}_3\text{-CoCo}$ , (b)  $\text{In}_2\text{Se}_3\text{-CoOs}$ , (c)  $\text{In}_2\text{Se}_3\text{-OsCo}$  and (d)  $\text{In}_2\text{Se}_3\text{-OsOs}$ .

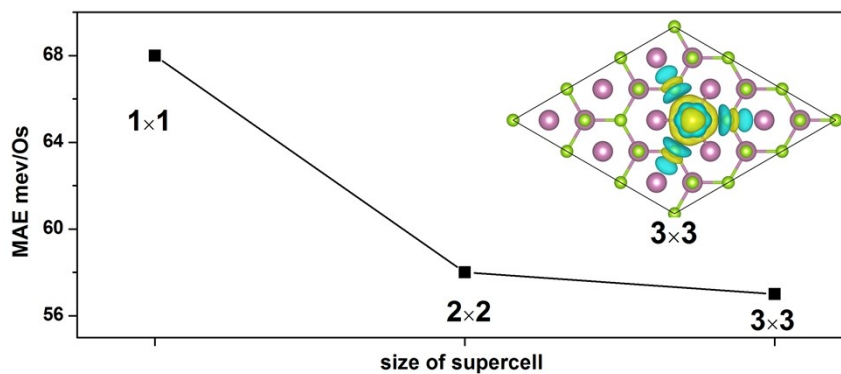


Fig. S2 Dependence of size of supercell on the MAE of Os atom in  $\text{In}_2\text{Se}_3\text{-CoOs}$  with FE polarization  $P\downarrow$ . Inset show the charge difference density of the  $3\times 3$   $\text{In}_2\text{Se}_3\text{-CoOs}$  supercell with FE polarization  $P\downarrow$ .

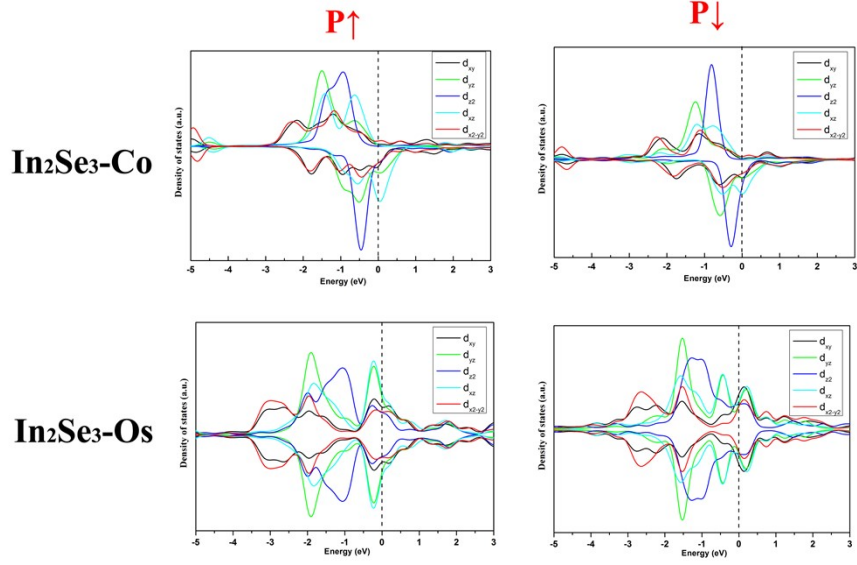


Fig. S3 PDOS of Co atom and Os atom in  $\text{In}_2\text{Se}_3\text{-Co}$  and  $\text{In}_2\text{Se}_3\text{-Os}$  with FE polarization  $P\uparrow$  and  $P\downarrow$ .

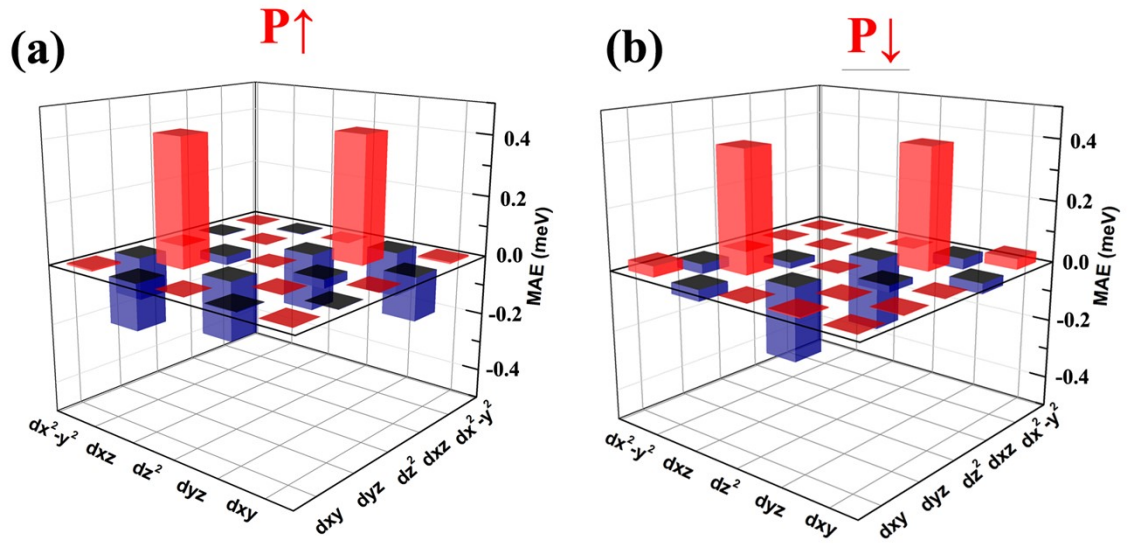


Fig. S4 Orbital-resolved MAE of Co atom in  $\text{In}_2\text{Se}_3\text{-Co}$  with FE polarization  $P\uparrow$  and  $P\downarrow$ .

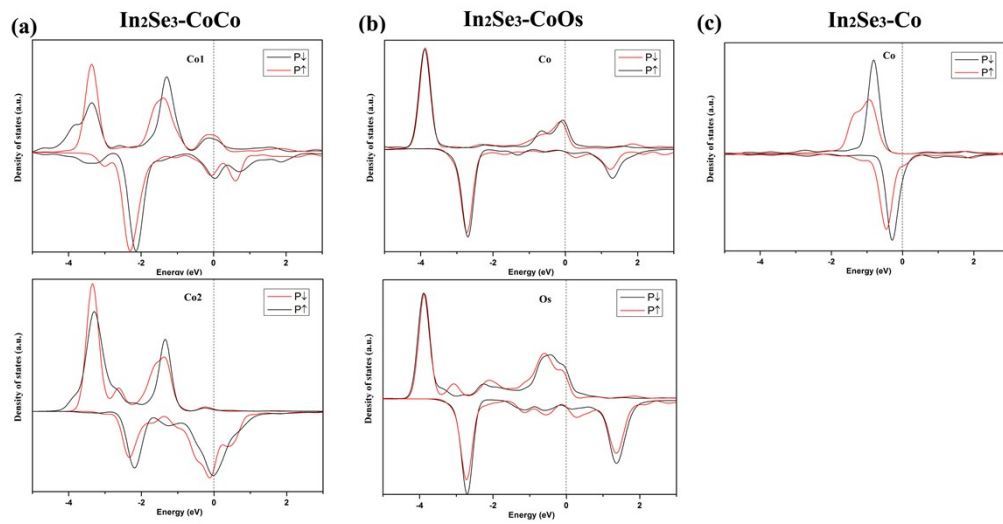


Fig. S5  $d_{z^2}$  PDOS of (a) Co1 and Co2 atoms in  $\text{In}_2\text{Se}_3\text{-CoCo}$ , (b) Co and Co atoms in  $\text{In}_2\text{Se}_3\text{-CoOs}$  and (c) Co atom in  $\text{In}_2\text{Se}_3\text{-Co}$  with FE polarization  $P_{\uparrow}$  and  $P_{\downarrow}$ .