Supplementary information for

Multiferroelectricity in two-dimensional Indium Pnictide optoelectronic materials

Jingwen Jiang[a], Zhuang Ma[*b], Yiguo Xu[*c]

[a] *School of Information Engineering, Jiangmen Polytechnic, Jiangmen, 529030, China*

[b] *School of Physics and Telecommunication Engineering, Zhoukou Normal University, Zhoukou, 466001, China*

[c] *Academy for Advanced Interdisciplinary Studies, Southern University of Science and Technology, Shenzhen, 518055, China*

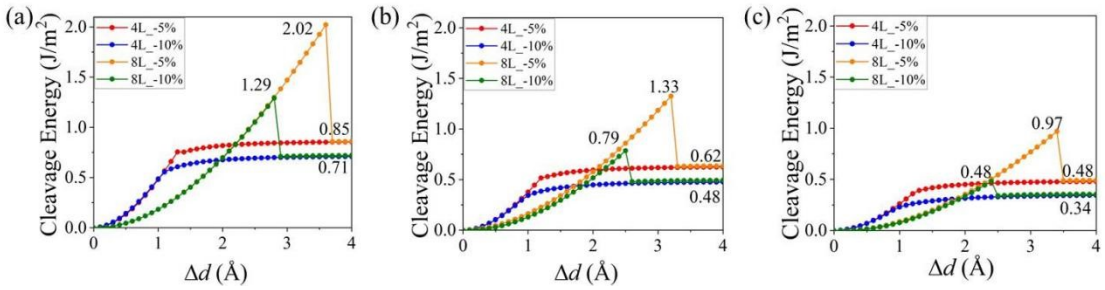

Fig. S1 Cleavage energy for indium pnictides with different strain.

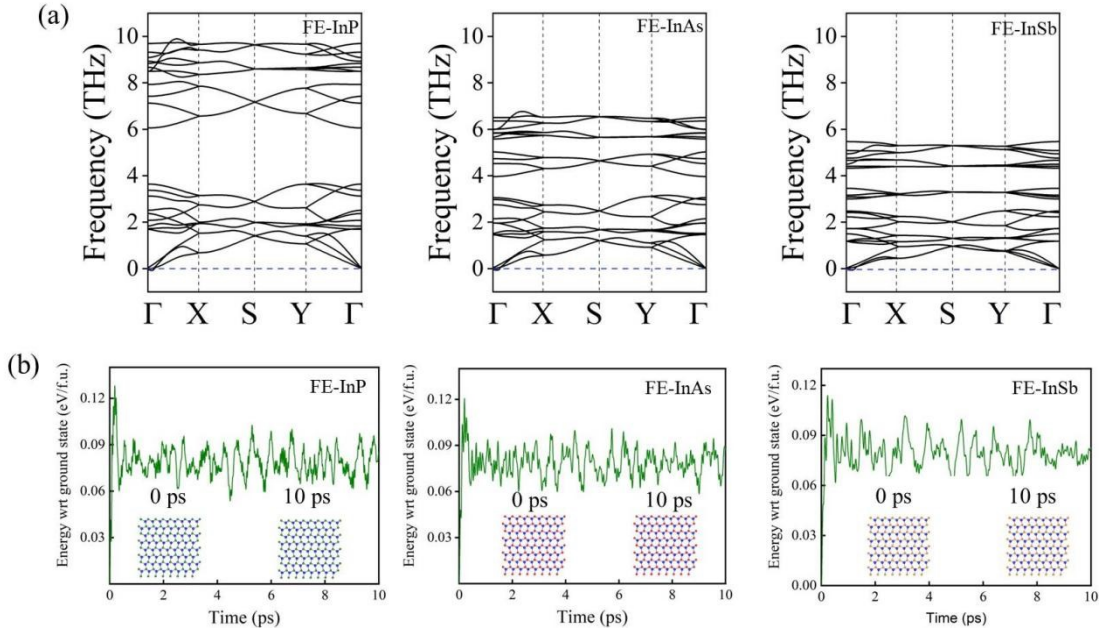

Fig. S2 (a) Phonon spectrum and **(b)** AIMD simulation for monolayer indium pnictides, which suggests that the monolayer structure is stable with non-imaginary frequency phonon dispersion and no phase change.

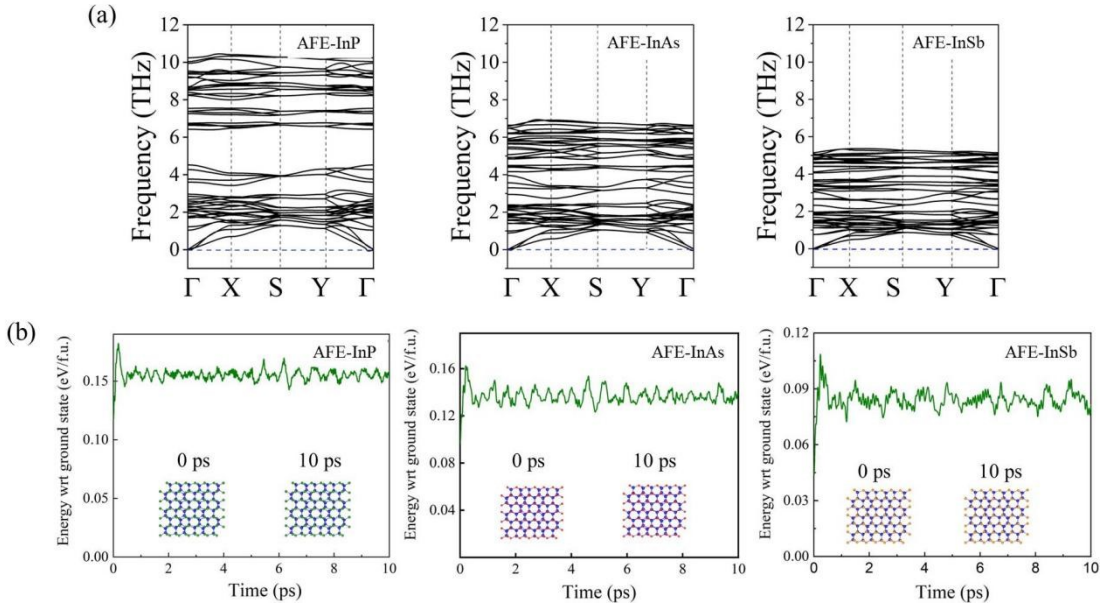

Fig. S3 (a) Phonon spectrum and **(b)** AIMD simulation for AFE indium pnictides, which suggest that the AFE structure is stable with non-imaginary frequency phonon dispersion and no phase change.

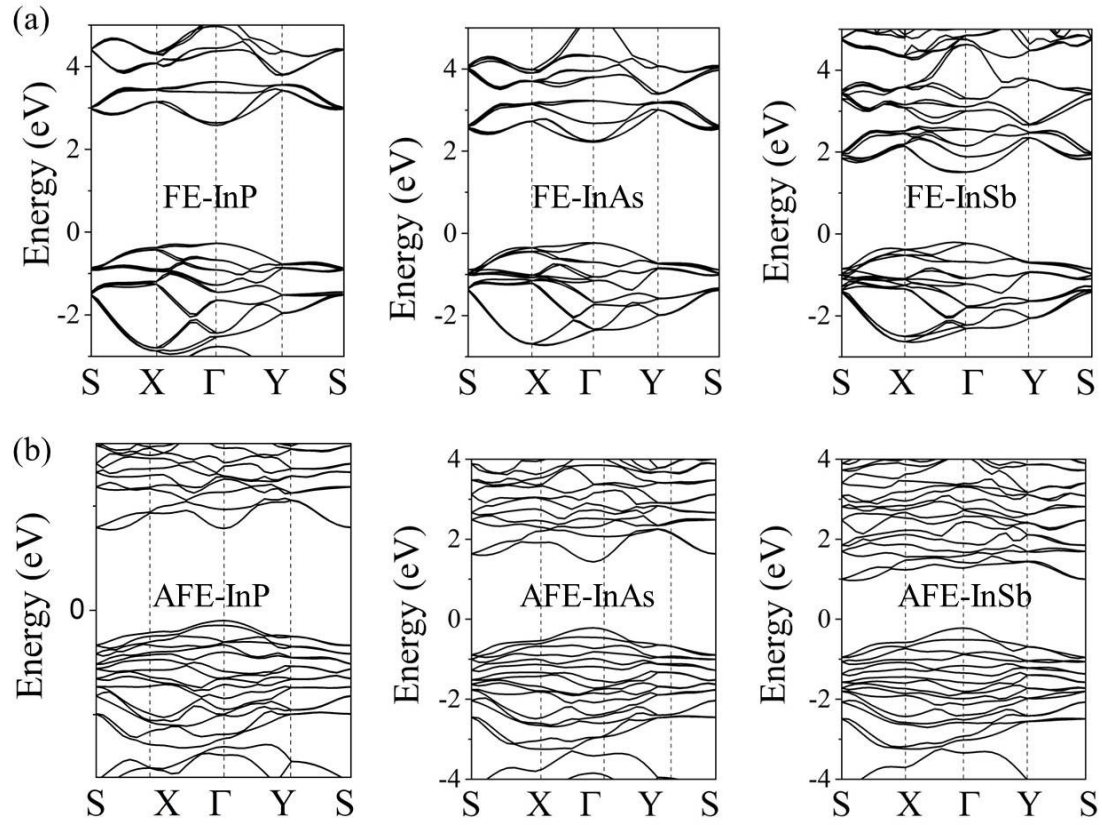

Fig. S4 Band structure of FE (a) and AFE (b) phases of indium pnictides from HSE06+SOC.

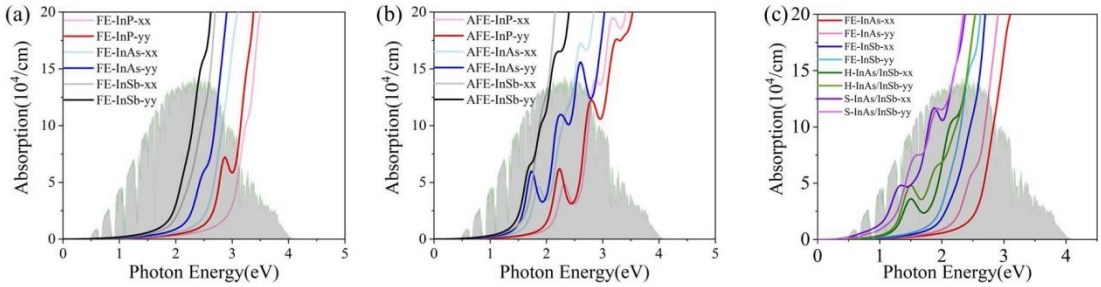

Fig. S5 Optical absorption coefficient of FE (a) and AFE (b) of indium pnictides, (c) heterojunction and superlattice of InAs/InSb phases. The reference air-mass 1.5-solar spectral irradiance is plotted in gray shadow.

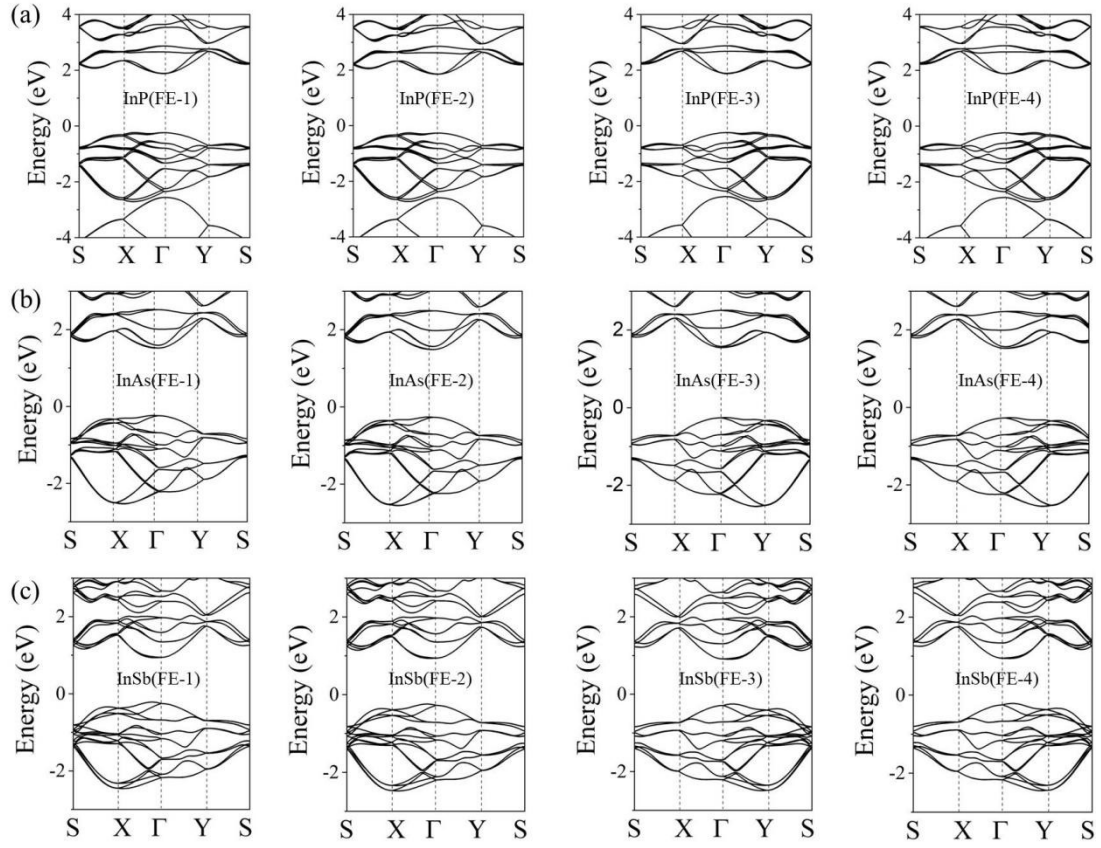

Fig. S6 Electronic band structures with SOC for indium pnictides monolayers with FE ($Pca2_1$) symmetry.

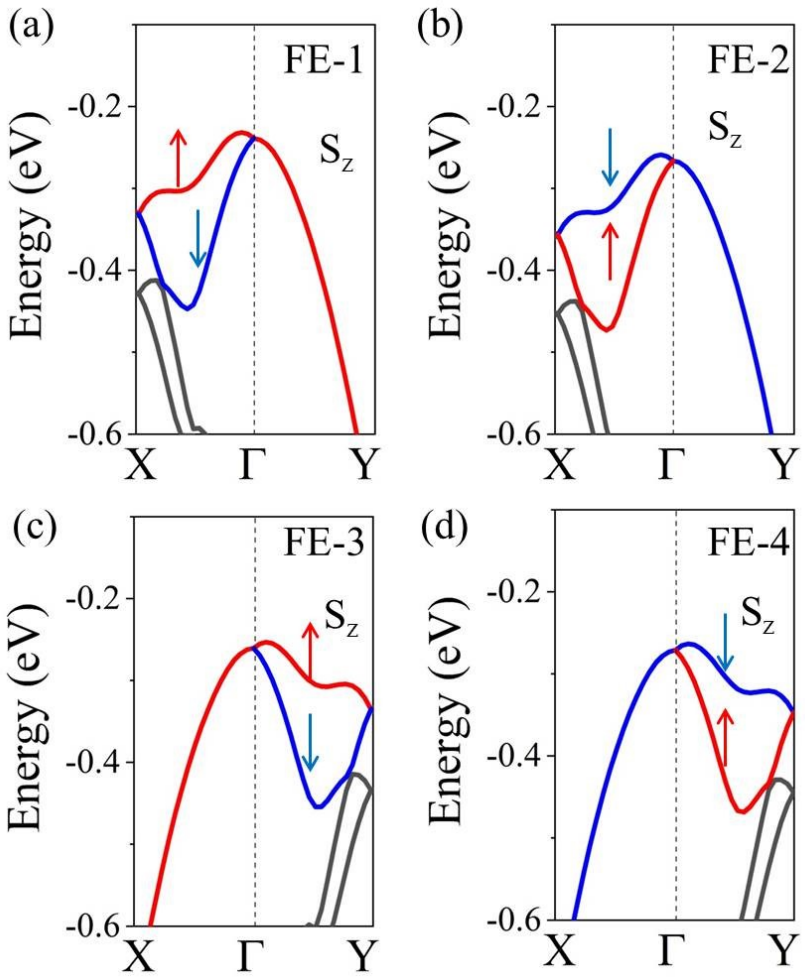

Fig. S7 The highlighted spin-split bands around VBM region of InAs monolayer. The splitting bands along Γ-Y/X (spin degenerate/splitting) and Γ-X/Y (spin degenerate/splitting) high-symmetry lines around VBM for $FE_{A/B}$ phases indicate the anisotropy flipping feature during the FC phase change process. The spin up and down property is inversed accompanied by the FE-1/2 phase change, in which the red and blue arrows indicate S_z (up) and $-S_z$ (down) spin orientation in the momentum space, respectively.

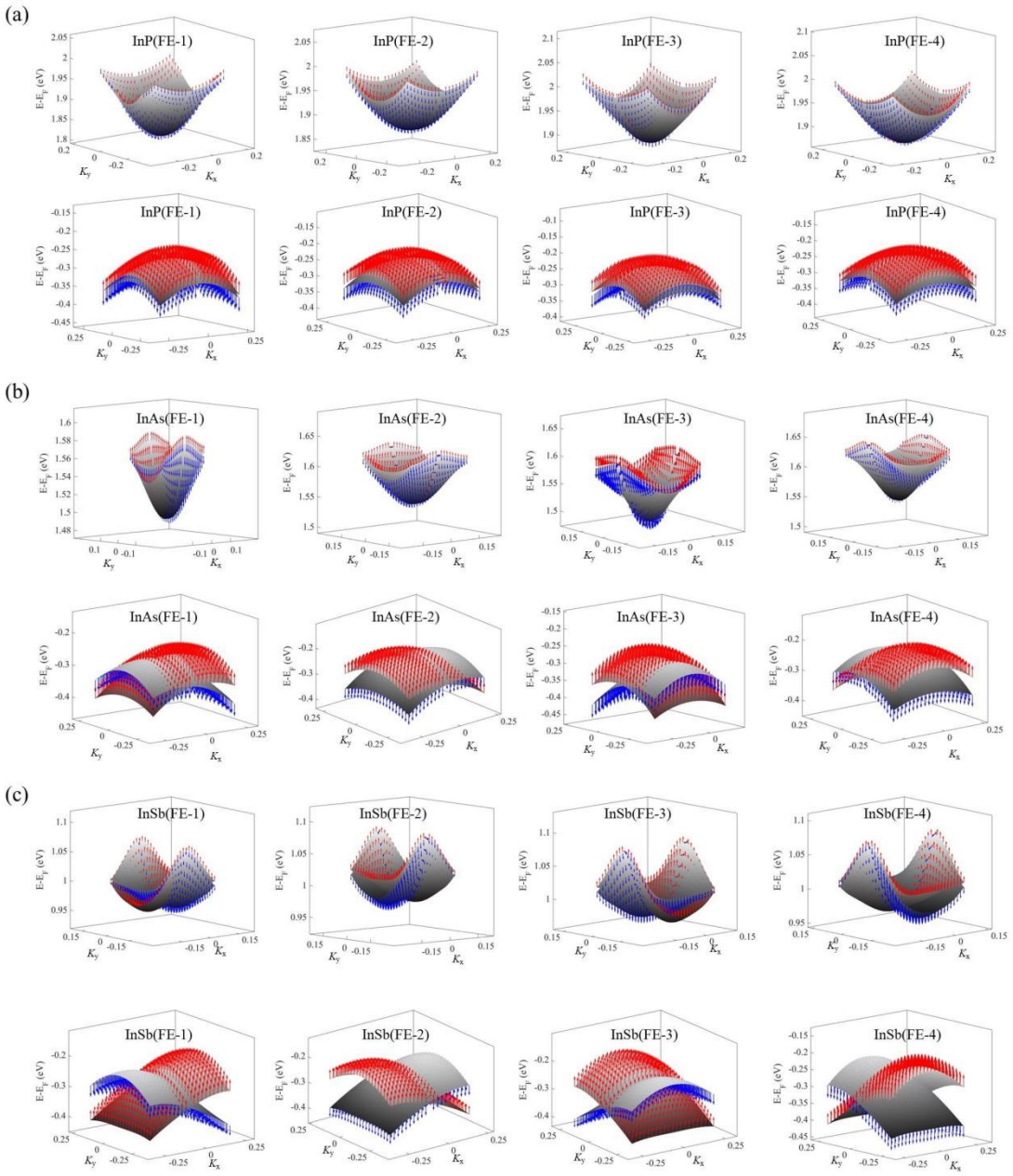

Fig. S8 (a-c) Switchable spin texture projected to the *k* space for the upper and lower bands of monolayer indium pnictides around CBM and VBM, which is expected by reversing the in-plane ferroelectric polarization.

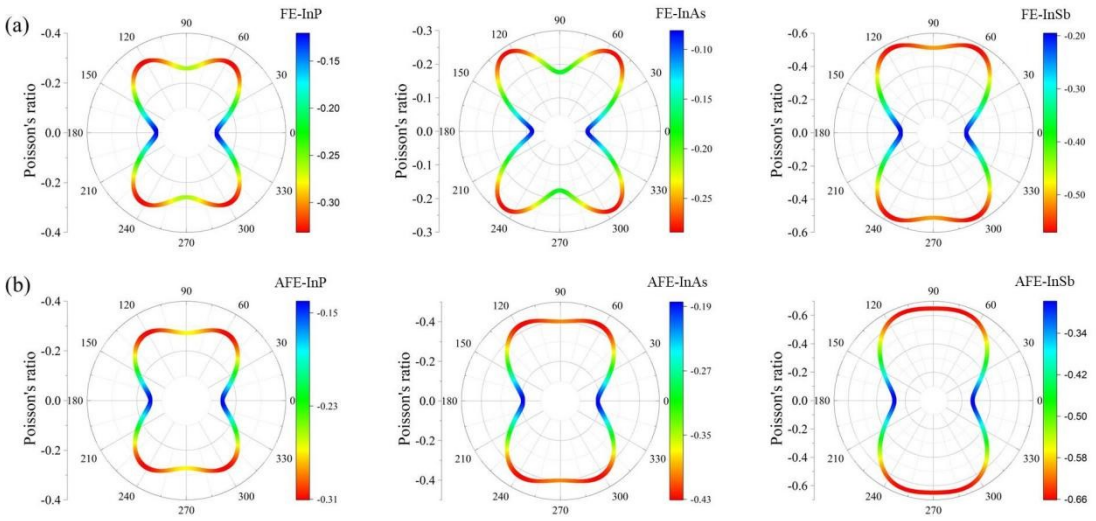

Fig. S9 Anisotropic in-plane NPR response for 2D FE (a) and AFE (b) indium pnictides.

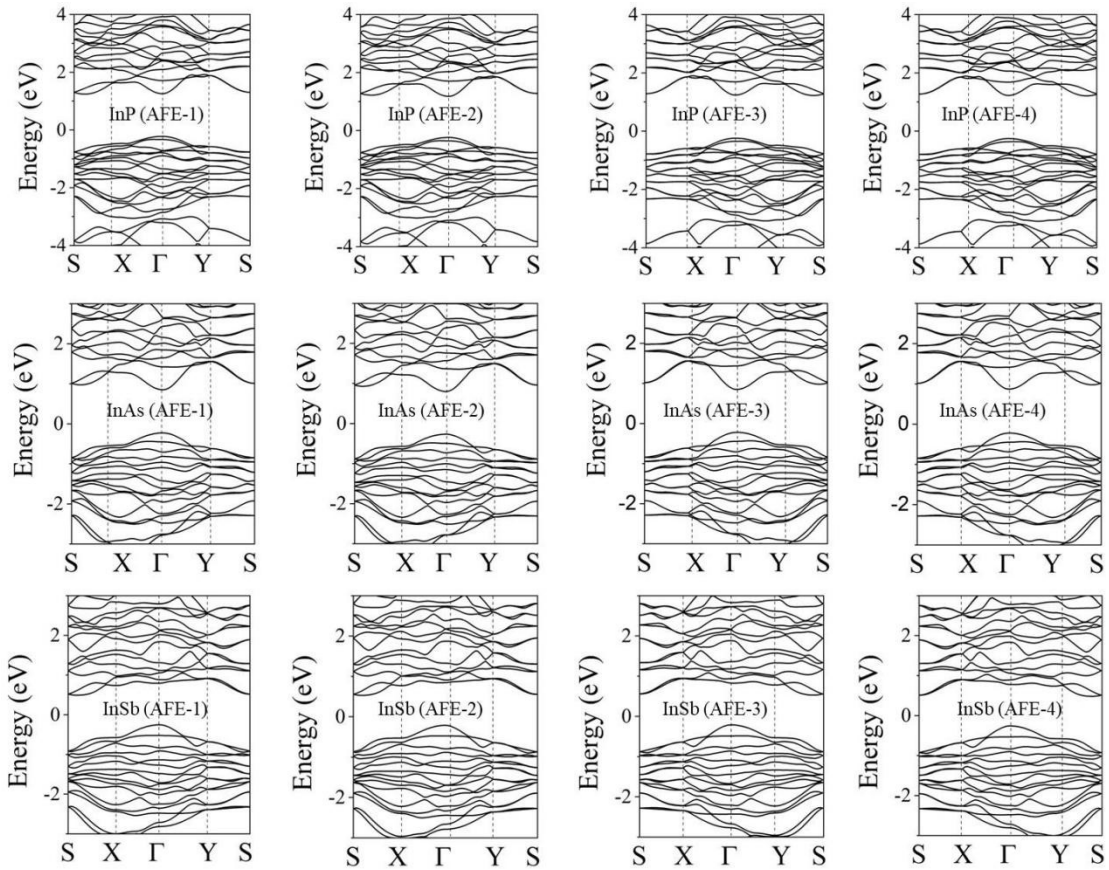

Fig. S10 Electronic band structures with SOC for indium pnictides with AFE (*P*2/*c*)

symmetry.

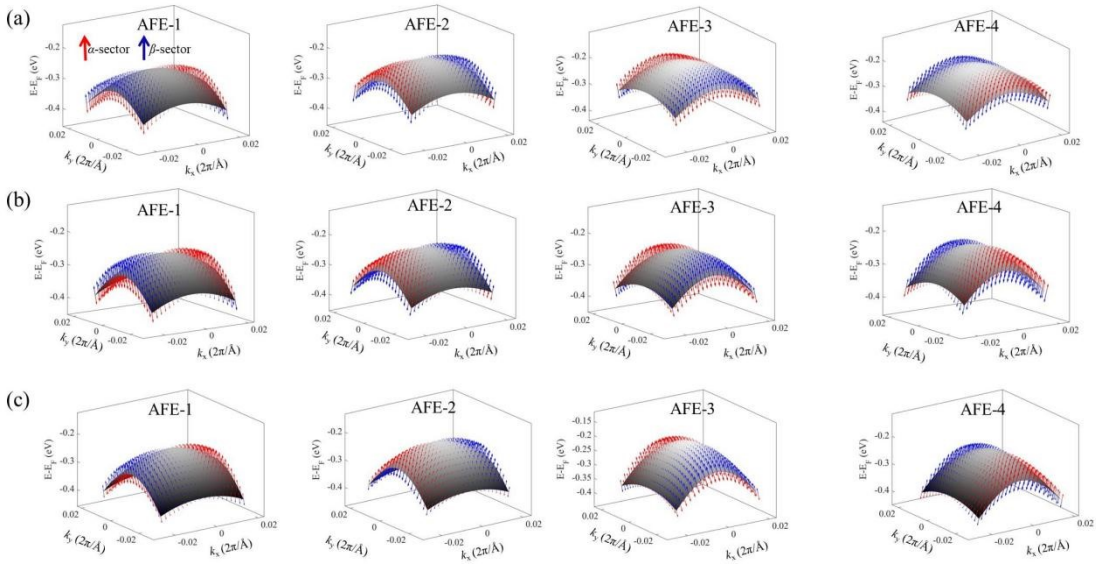

Fig. S11 (a-c) Switchable hidden spin texture projected to the *k* space for the upper and lower bands of AFE indium pnictides around VBM.