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New monolayer ternary In-containing sesquichalcogenides $BiInSe_3$, $SbInSe_3$, $BiInTe_3$, and $SbInTe_3$ with high stability and extraordinary piezoelectric properties

Huabing Yin,[a,b] Guang-Ping Zheng,[*b] Yuanxu Wang,[†a] and Bingjian Yao[c]

[a.] *Institute for Computational Materials Science, School of Physics and Electronics, Henan University, Kaifeng 475004, China.*

[b.] *Department of Mechanical Engineering, The Hong Kong Polytechnic University, Hong Kong 999077, China.*

[c.] *College of Chemistry, Chemical Engineering and Materials Science, Collaborative Innovation Center of Functionalized Probes for Chemical Imaging in Universities of Shandong, Shandong Normal University, Jinan 250014, China*

E-mails: [*]mmzheng@polyu.edu.hk (G.-P. Zheng); [†]wangyx@henu.edu.cn (Y. Wang)

Computational details for piezoelectric coefficients using VASP code.

According to Eqn (1) and (2) in the main text, we obtain the piezoelectric coefficients e_{ijk} and d_{ijk} from the polarization tensor P_i, strain tensor ε_{jk}, and stress tensor σ_{jk}. The indices i=1, 2, and 3 for polarization tensor P_i indicate the polarization components along x-, y-, and z-directions, respectively. In Voigt notation, for ε_{jk} and σ_{jk} tensors, the indices j and k can also be labeled as 1 = xx, 2 = yy, 3 = zz, 4 = yz, 5 = zx, and 6 = xy. However, in 2D materials, we only consider the in-plane strains and stresses, such as those with 1 (xx), 2 (yy), and 6 (xy) components. Therefore, the second-rank ε_{jk} tensors of quasi-2D systems under study can be indicated as only three forms, i.e., ε_{11}, ε_{22}, and ε_{12}, where 1 or 2 corresponds to x- or y-direction as defined in Figure 1 in the main text and the relationship of $\varepsilon_{12} = \varepsilon_{21}$ could exist. Therefore, the 2D elastic and piezoelectric tensors become simplified forms of C_{ij} and e_{ij}. In the DFT calculations performed using VASP code, we renormalize the 2D coefficients C_{ij}^{2D} and e_{ij}^{2D} by multiplying the lattice parameter along the z-direction because of the existence of vacuum space in the direction that is perpendicular to the 2D layer.

The modern theory of polarization based on the Berry's phase approximation, which has been proved to be the most effective method in evaluating the piezoelectricity of 2D materials, are applied to calculate relaxed-ion piezoelectric coefficients, e_{ij}, including both the ionic and electronic contributions. By definition, we perform the polarization vector calculations by using the **LCALCPOL** tag in VASP code. Once we obtain the polarization changes under a very small strain range, a linear fitting method is used to calculate the piezoelectric coefficients e_{11} and e_{31}, which reflect the polarization changes ΔP_1 along the x-direction and ΔP_3 along the z-direction under the in-plane uniaxial strains ε_{11} along the x-direction, respectively. Because of the noncentrosymmetric C_{3v} point group symmetry of these four monolayer ternary compounds, $BiInSe_3$, $SbInSe_3$, $BiInTe_3$, and $SbInTe_3$, only the piezoelectric e_{11} and e_{31} coefficients are non-zero and independent. As described in Eqn (3) and (4) in the main text, the relationships of $e_{12} = -e_{11}$, $e_{32} = e_{31}$, and $e_{26} = -\frac{e_{11}}{2}$ could exist.

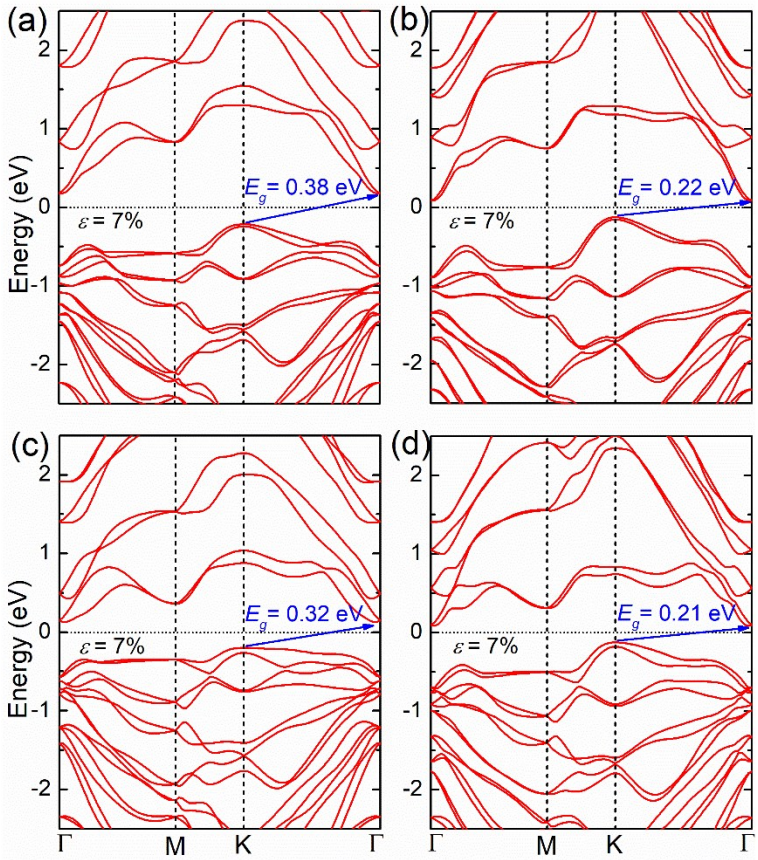

Fig. S1 PBE+SOC band structures of (a) $BiInSe_3$, (b) $SbInSe_3$, (c) $BiInTe_3$, and (d) $SbInTe_3$ ternary monolayers under an in-plane 7% tensile strain. The strain is defined as $\varepsilon = (a - a_0)/a_0$, where a_0 is the lattice constant in plane and a is that under external strain. The band gaps are highlighted in blue. The Fermi level is at 0 eV.

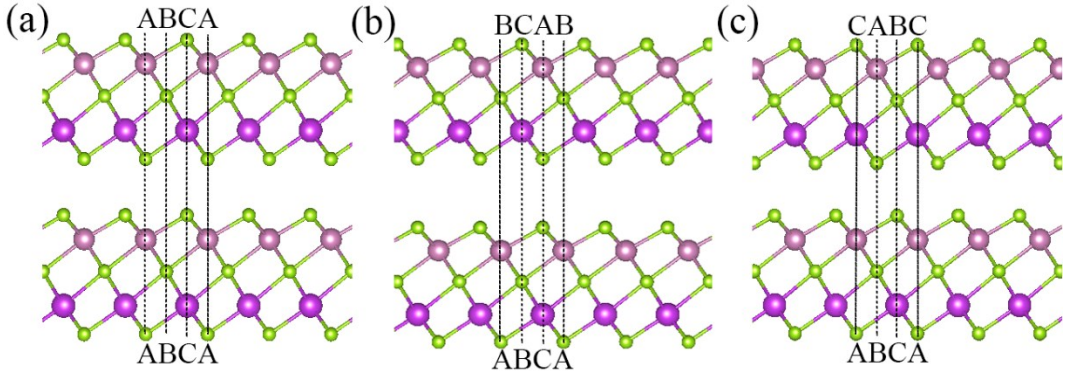

Fig. S2 Three types of binary stackings: (a) AA, (b) AB, and (c) AC. The atomic layers in one unit cell are illustrated as A, B, and C.

Table S1 Calculated averaged relaxed-ion piezoelectric coefficients e_{11}/N and e_{31}/N of bilayer $BiInTe_3$ with three different stacking models. N is the layer number. The coefficient is in unit of 10^{-10} C/m.

	stacking models	*N*	e_{11}/N	e_{31}/N
monolayer	A	1	22.83	1.55
bilayer	AA	2	22.42	1.44
	AB	2	22.94	1.53
	AC	2	22.75	1.57