Electronic Supplementary Information

Ultrahigh mechanical flexibility induced superior piezoelectricity of InSeBr-type 2D Janus materials

Xiaobo Shi,[ab] Shujuan Jiang,[ac] Xianwei Han,*[a] Min Wei,[d] Bing Wang, [a] Gaofeng Zhao,*[a] Guang-Ping Zheng*[c] and Huabing Yin*[a]

[a]*Institute for Computational Materials Science, International Joint Research Laboratory of New Energy Materials and Devices of Henan Province, School of Physics and Electronics, Henan University, Kaifeng 475004, China*

[b]*Institute of Artificial Intelligence, Henan Finance University, Zhengzhou 450046, China.*

[c]*Department of Mechanical Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong 999077, China*

[d]*Department of Physics and Electronic Engineering, Jinzhong University, Jinzhong 030619, China*

E-mails: xianweihan@henu.edu.cn, 10110094@vip.henu.edu.cn, mmzheng@polyu.edu.hk, and yhb@henu.edu.cn

Table S1. The optimized lattice constants of bulk InSeBr-type materials calculated by different vdW correction methods, including D2 method of Grimme (DFT-D2),[1] zero damping D3 method of Grimme (DFT-D3),[2] D3 method of Grimme with Becke-Jonson damping (DFT-D3+BJ),[3] Tkatchenko-Scheffler method (DFT-TS).[4] And the corresponding experiment data are also listed for comparison as well. DFT-D2 method can effectively reproduce the experiment data.

Material	DFT-D2	DFT-D3	DFT-D3+BJ	DFT-TS	Exp.[5, 6]
InSCl	a=3.76 Å, c=17.91 Å	a=3.82 Å, c=17.81 Å	a=3.80 Å, c=17.70 Å	a=3.84 Å, c=17.98 Å	a=3.72 Å, c=17.95 Å
InSBr	a=3.81 Å, c=18.51 Å	a=3.86 Å, c=18.53 Å	a=3.85 Å, c=18.46 Å	a=3.89 Å, c=18.82 Å	a=3.83 Å, c=18.75 Å
InSeCl	a=3.88 Å, c=18.18 Å	a=3.94 Å, c=18.12 Å	a=3.92 Å, c=18.09 Å	a=3.97 Å, c=18.38 Å	a=3.85 Å, c=18.44 Å
InSeBr	a=3.92 Å, c=18.94 Å	a=3.98 Å, c=18.92 Å	a=3.96 Å, c=18.84 Å	a=4.01 Å, c=19.25 Å	a=3.92 Å, c=19.01 Å

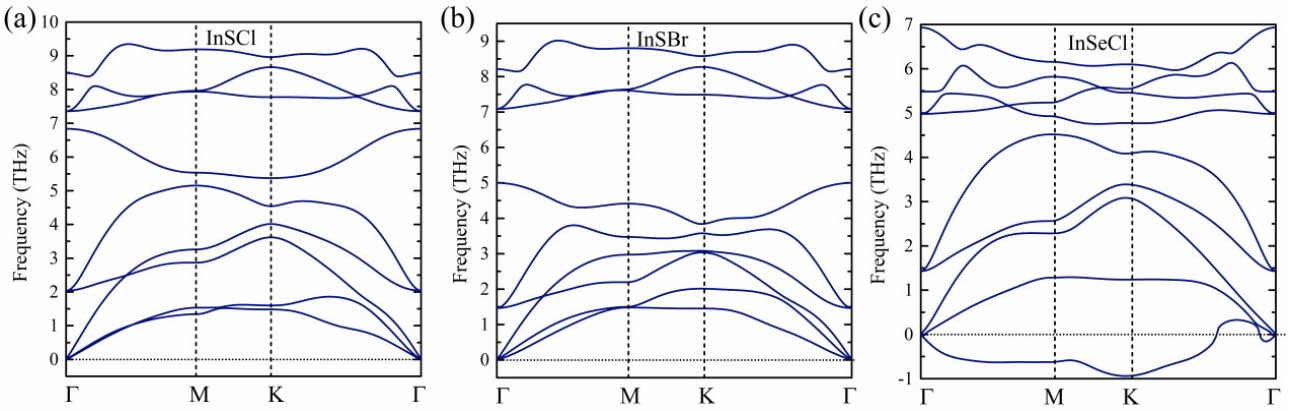

Fig. S1. Phonon spectra of (a) InSCl, (b) InSBr, and (c) InSeCl monolayers.

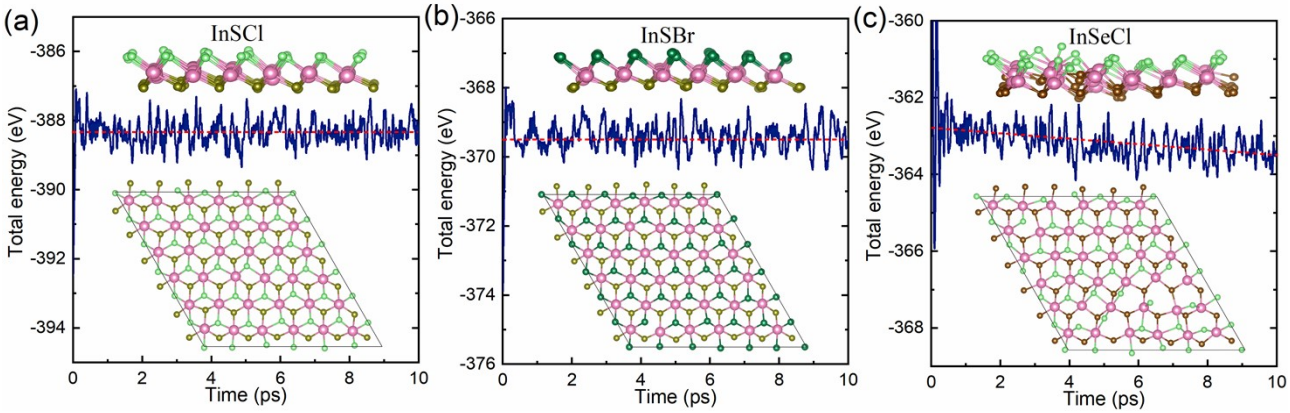

Fig. S2. Evolutions of atomic structures and potential energies of Janus (a) InSCl, (b) InSBr, and (c) InSeCl monolayers during the AIMD simulations at 300K for 10 ps. The red line indicates the average total energy during the simulations. For InSCl and InSBr monolayers, the initial equilibrium nanostructures are well maintained and the total energy has small fluctuation during the AIMD simulation at 300 K, suggesting that their good thermal stability. For InSeCl monolayer, the obvious

broken bonds and reduction of total energy indicate that it is thermally unstable.

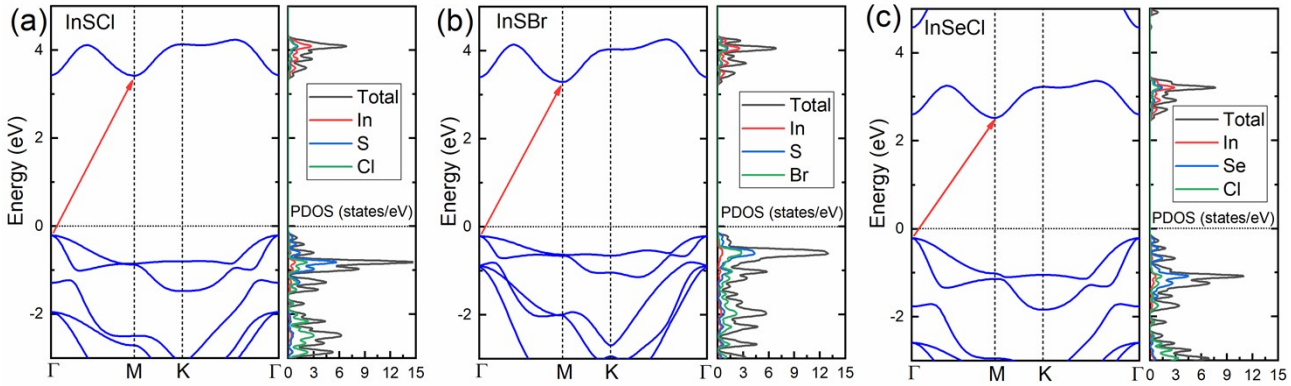

Fig. S3. Electronic band structures and partial density of states (PDOS) of Janus (a) InSCl, (b) InSBr, and (c) InSeCl monolayers calculated by HSE06 functional. The red arrow indicates the positions of the VBM and CBM, and the Fermi level is set at zero.

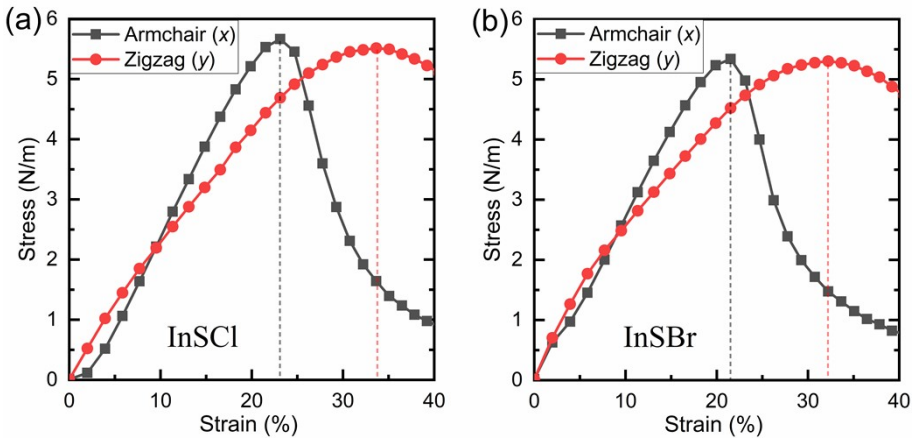

Fig. S4. The calculated stress-strain relations of Janus (a) InSCl and (b) InSBr monolayers under tensile deformation along armchair (*x*-) and zigzag (*y*-) directions.

Table S2. The comparison of piezoelectric strain coefficients between InSeBr-type monolayers and other reported 2D monolayers and 3D perovskite-based materials.

Material	Piezoelectric strain coefficient
2D monolayer systems: d_{11} (pm/V)	
InSCl	149~154
InSBr	51~53
InSeBr	2361~3224
SnSe	251
SbAs	243
$BiInTe_3$	362
MoS_2	3.73
Janus $GaInS_2$	8.33
Janus SbTeI	12.95
3D perovskite-based systems: d_{33} (pm/V)	
Pb(Zr, Ti)O_3 (PZT)	360~900
$PbTiO_3$ (PT)	1000~2000
$K_{0.5}Na_{0.5}NbO_3$ (KNN)	355~1030
$Bi_{0.5}Na_{0.5}TiO_3$-$Bi_{0.5}K_{0.5}TiO_3$ (BNKT)	250~1400
$Bi_{0.5}Na_{0.5}TiO_3$ (BNT)	249~1100
$Ba(Zr_{0.2}Ti_{0.8})O_3$-x$(Ba_{0.7}Ca_{0.3})TiO_3$ (BZT-BCT)	400~1310
$(Ba_{0.94}Ca_{0.06})$-$(Ti_{0.95}Zr_{0.05})O_3$ (BCTZ)	2027

Table S3. The in-plane elastic stiffness coefficients C_{11} and C_{12} of the 2D Janus InSeBr-type monolayers. The superscripts of *el* and *ion* represent electronic and ionic contributions, respectively. $C_{11} = C_{11}^{el} + C_{11}^{ion}$ and $C_{12} = C_{12}^{el} + C_{12}^{ion}$.

Material	C_{11}^{el}	C_{12}^{el}	C_{11}^{ion}	C_{12}^{ion}	C_{11}	C_{12}
InSCl	62.20	25.57	-27.98	1.98	34.22	27.55
InSBr	60.57	24.32	-21.24	-1.11	39.33	23.21
InSeCl	53.78	21.89	-40.42	17.94	13.36	39.83
InSeBr	52.65	20.78	-25.71	5.75	26.94	26.53

References

1. S. Grimme, *J. Comput. Chem.*, 2006, **27**, 1787-1799.
2. S. Grimme, J. Antony, S. Ehrlich and H. Krieg, *J. Chem. Phys.*, 2010, **132**, 154104.
3. S. Grimme, S. Ehrlich and L. Goerigk, *J. Comput. Chem.*, 2011, **32**, 1456-1465.
4. A. Tkatchenko and M. Scheffler, *Phys. Rev. Lett.*, 2009, **102**, 073005.
5. H. Hahn and W. Nickels, *Z. Anorg. Allg. Chem.*, 1962, **314**, 307.

6. R. Kniep and W. Welzel, *Zeitschrift für Naturforschung B*, 1985, **40**, 26-31.