Supplementary Information：

Robust Ferroelectricity in Two-dimensional SbN and BiP

Chang Liu,[1,2] *Wenhui Wan,*[*,1] *Jie Ma,*[1] *Wei Guo,*[1] *and Yugui Yao*[*,1]

[1]Beijing Key Laboratory of Nanophotonics and Ultrafine Optoelectronic Systems, School of Physics, Beijing Institute of Technology, Beijing 100081, China

[2]Kunming Institute of Physics, Kunming, 650223, China

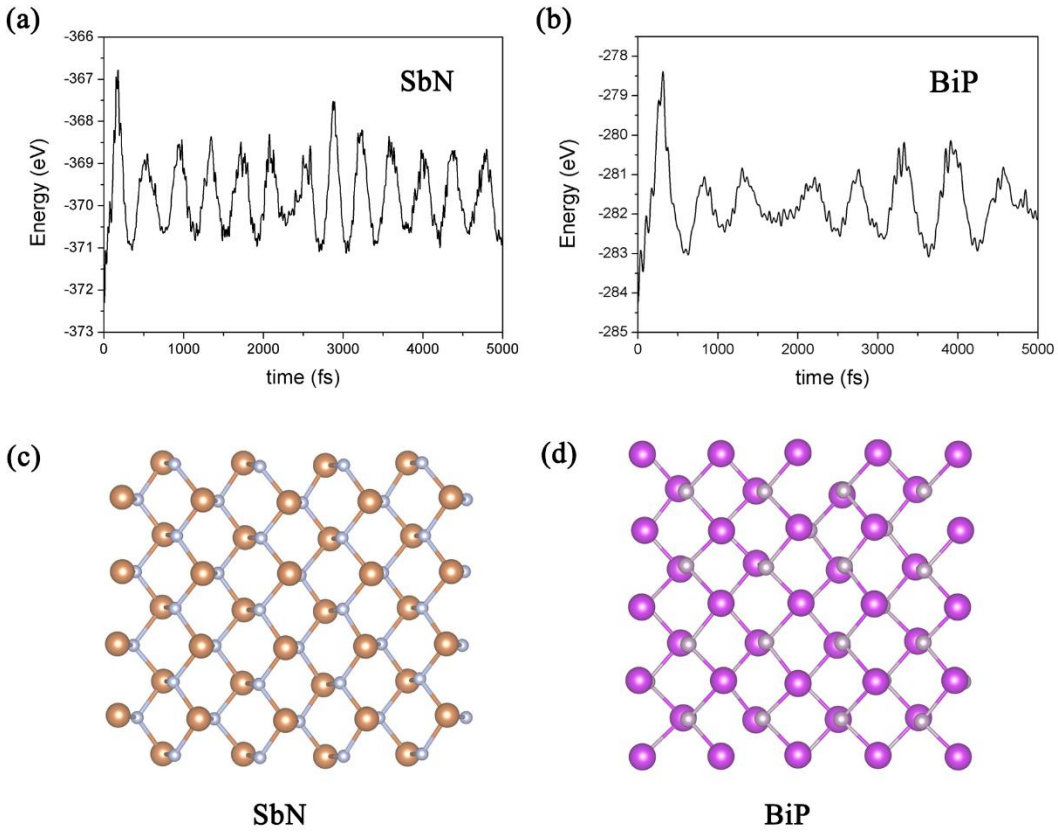

Fig. S1 Relationships between total energy and time during 300K MD simulations of (a) SbN and (b) BiP, respectively. The MD process is imposed for 5 ps time scale with 2500 steps (2fs/step). The final geometric structures of (c) SbN and (d) BiP at the end of 5 ps are also shown.

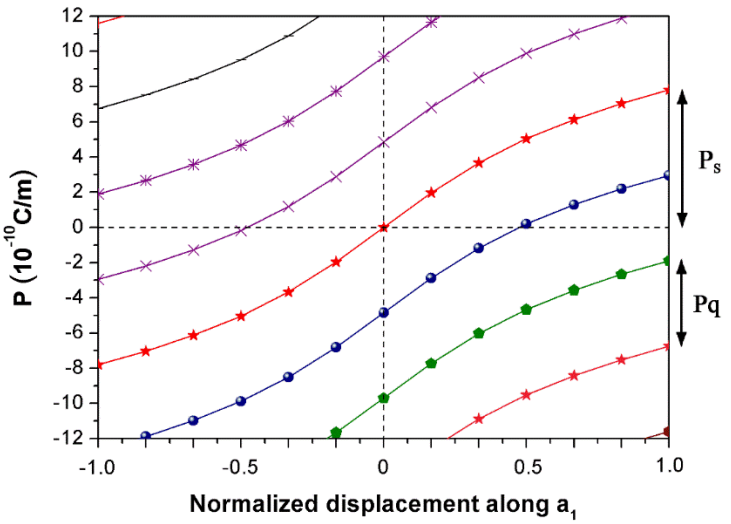

Fig. S2 Calculated total polarization of SbN as a function of normalized displacement where the centrosymmetric paraelectric phase (0% displacement) is at the center, and two ferroelectric ground states are at two ends. P_s represents the spontaneous polarization and Pq represents the polarization quanta.

Table S1 The spontaneous polarization P_s $(10^{-10}\mathrm{C/m})$ and switching barrier E_b (meV/unit cell) of SbN and BiP, respectively.

compound	$P_s\,(10^{-10}\mathrm{C/m})$	E_b (meV)
SbN	7.81	604
BiP	5.35	65

Details for Monte Carlo simulations:

We used the Landau theory with the polarization P as the order parameter. The system free energy is written as a Taylor expansion in terms of the polarization: $\mathrm{F} = \sum_i (\frac{A}{2}P_i^2 + \frac{B}{4}P_i^4 + \frac{C}{6}P_i^6) + \frac{D}{2}\sum_{<i,j>}(P_i - P_j)^2$, where P_i is the polarization of each unit cell, < > denotes the nearest neighbor. As we use the mean-field approximation, only the nearest neighbor has been taken into consideration (Ref. 7). The first three terms are associated with the energy contribution from the

local modes up to the sixth order. The last term captures the coupling between the nearest local modes, which include the 2D geometrical character and are crucial for the ordering and the phase transition. After we obtained the value of parameters by fitting, this model are used in the Monte Carlo simulations to investigate the temperature effects and the phase transition. The fitted values of the parameters are shown in **Table S2.**

Table S2 Fitted parameters in the equation: $\mathrm{F} = \sum_i (\frac{A}{2} P_i^2 + \frac{B}{4} P_i^4 + \frac{C}{6} P_i^6) + \frac{D}{2} \sum_{<i,j>} (P_i - P_j)^2$. The unit of P_i is 10^{-10}C/m, and the unit of F is meV.

material	A	B	C	D
SbN	-34.315	-0.465	0.015	5.203
BiP	-5.507	-1.588	0.078	1.579

Table S3 Energy difference (meV/unit cell) between AA-stacking and other bilayer stacking order of SbN and BiP. Considering the Van der Waals interaction between layers, both optB88-vdW functional and vdW-D3 functional were respectively imposed in the calculations. After structural relaxation, for both SbN and BiP, the anti-AB structure transforms to anti-AA while the anti-AD transform to anti-AC stacking.

Stacking type	SbN		BiP	
	optB88	vdW-D3	optB88	vdW-D3
AA	0	0	0	0
AB	66	67	150	220
AC	-7	-19	104	184
AD	124	116	302	420
anti-AA	8	-1	79	171
anti-AC	18	14	120	195