Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information

Symmetry-breaking Induced Large Piezoelectricity in Janus Tellurene Materials

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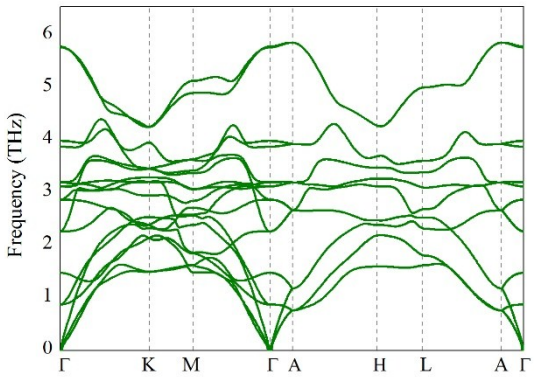

Figure S1. Phonon dispersion of the Janus Te2Se multilayer structure (d). The high symmetry k points are: *Γ* (0, 0, 0), *K* (-1/3, 2/3, 0), *M* (0, 1/2, 0), *A* (0, 0, 1/2), *H* (-1/3, 2/3, 1/2), *L* (0, 1/2, 1/2).

Table S1. Comparisons of clamped-ion elastic stiffness constants (C_{ij}) and the clamped-ion piezoelectric coefficients (e_{ij}/d_{ij}) in unit of N/m, 10^{-10} C/m, and pm/V, respectively.

Materials	C_{11}	C_{12}	e_{11}	e_{31}	d_{11}	d_{31}
Janus Te_2Se	57.92	15.33	3.994	0.146	9.378	0.199
α-Tellurene	53.4	15.0	—	—	—	—
MoS_2 [1]	153	48	3.06	—	2.91	—
Janus In_2SSe[2]	88	27	7.94	0.07	13.06	0.11

Table S2. Relative energy per unit cell (ΔE), lattice parameters (a/c) and the unit-cell volume (V) of the unit cell of Janus Te_2Se multilayers with the structures shown in Figure 6. The units of ΔE, a/c, and V are meV, Å, and (Å^3), respectively.

Structure	ΔE	a	c	V
(a)	0	4.157	12.115	181.33
(b)	208	4.099	13.573	197.52
(c)	315	4.091	14.267	206.76
(d)	47	4.130	12.504	184.71
(e)	68	4.118	12.688	186.33

Table S3. The clamped-ion elastic stiffness coefficients (C_{ij}) and the clamped-ion piezoelectric coefficients (e_{ij}/d_{ij}) of the Janus Te_2Se multilayer with five structures in Figure 6. The units of C_{ij}, e_{ij} and d_{ij} are GPa, 10^{-10} C/m and pm/V, respectively.

Structure	C_{11}	C_{12}	C_{13}	C_{33}	e_{11}	e_{31}	e_{33}	d_{11}	d_{31}	d_{33}
(a)	88.08	22.78	46.23	89.10	0.755	0.170	-0.428	11.562	6.232	-11.269
(b)	81.53	22.85	37.22	83.92	0.601	0.098	-0.177	10.248	2.473	-4.308
(c)	78.90	21.59	34.02	80.52	0	0.089	-0.157	0	2.163	-3.777
(d)	82.95	26.59	44.65	85.20	0	0.076	-0.178	0	2.709	-4.934
(e)	85.20	23.19	42.43	84.45	0	0.143	-0.322	0	4.637	-8.469

Reference

1. K.-A. N. Duerloo, M. T. Ong and E. J. Reed, *The Journal of Physical Chemistry Letters*, 2012, **3**, 2871-2876.
2. Y. Guo, S. Zhou, Y. Bai and J. Zhao, *Applied Physics Letters*, 2017, **110**, 163102.