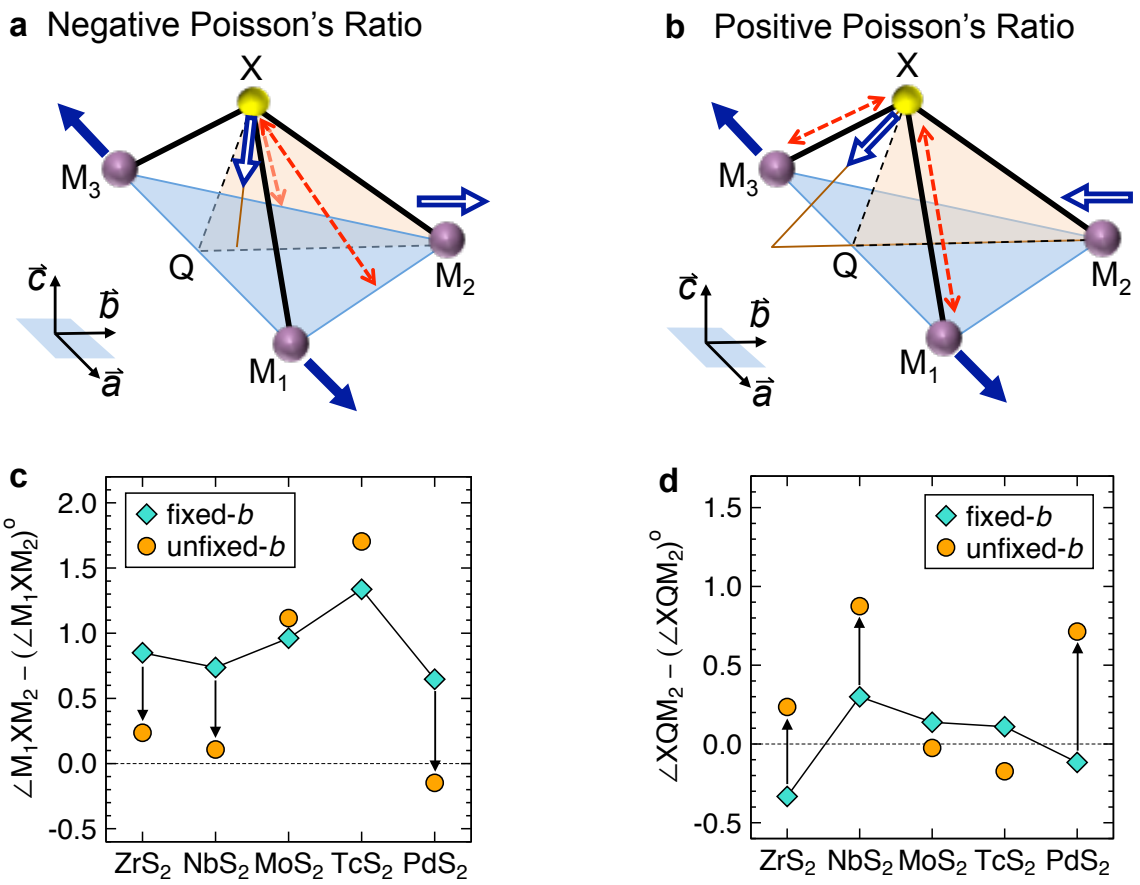

Supplementary Figure 1 | Deformation Mechanism. a-b, Motion of atoms in the auxetic and non-auxetic 1T-MX_2 compounds. The solid and open blue arrows indicate the applied force direction and resulting atom motion direction, respectively. The dashed red arrow indicates the direction of the t_{2g}-p orbital coupling force. **c,** Change in $\angle M_1XM_2$ after step-ii relaxation (i.e., the relaxation with the lattice *b* fixed to its value in the relaxed strain-free structure). **d**, Change in X-M_3-M_1-M_2 dihedral angle $\angle QXM_2$ $\angle M_1XM_2$ after step-iii relaxation. The $(\angle QXM_2)^o$ and $(\angle M_1XM_2)^o$ are the reference angles taken in the relaxed structure of corresponding strain-free MX_2. The diamond and circle symbols represent the angles extracted from the structures relaxed under fixed lattice-*b* and unfixed lattice-*b*, respectively, subjected to a 5% tension strain in the lattice-*a* direction. The solid lines and arrows in c-d are guides to the eyes.

Supplementary Table 1: The Poisson's ratios and lattice constants of groups 6-7 MX_2 compounds calculated from SCAN and HSE06. Available experimental lattice constants are shown in parentheses.

	In-plane Poisson's ratio along *a*: SCAN/HSE06			Lattice Constant *a* (Å): SCAN/HSE06 (Experiment)		
M^{4+} \ X_2	$-S_2$	$-Se_2$	$-Te_2$	$-S_2$	$-Se_2$	$-Te_2$
Mo	-0.07/**0.01**	-0.12/-0.00	-0.09/-0.01	**3.1998/3.1657 (3.201)**[1]	**3.2685/3.2529 (3.270)**[1]	3.4970/3.4626 (3.469)[2]
W	-0.04/**0.05**	-0.19/-0.05	-0.20/-0.06	**3.1908/3.1843 (3.222)**[1]	3.2574/3.2645	**3.4970/3.4759 (3.496)**[3]
Tc	-0.10/**0.03**	-0.04/**0.01**	-0.37/-0.02	3.0692/3.0255	3.1543/3.1386	3.4149/3.3486
Re	-0.13/-0.02	-0.03-0.02	-0.22/-0.09	3.0750/3.0590	3.1311/3.1381	3.3834/3.3551

Supplementary Table 2. SCAN-calculated lattice constants of 42 1T-MX_2 compounds. Available experimental values are shown in parentheses. Note the experimental lattice constants listed for d^5-d^6 MTe_2 compounds are extracted from the distorted 1T structure. The small difference in lattice constant between the undistorted and distorted phases is expected.

		Lattice Constant *a* (Å)		
M^{4+} \ X_2		-S_2	-Se_2	-Te_2
d^0	Ti	3.4055 (3.405)[4]	3.5165 (3.535)[4]	3.7648 (3.766)[4]
	Zr	3.6833 (3.662)[4]	3.7815 (3.770)[4]	4.0064 (3.950)[4]
	Hf	3.6153 (3.635)[4]	3.7180 (3.748)[4]	3.9606 (3.949)[5]
d^1	V	3.2668 (3.29)[6]	3.3260(3.352)[4]	3.6022 (3.595)[4]
	Nb	3.3870	3.4845	3.6738 (3.642)[7]
	Ta	3.3524 (3.346)[4]	3.4602 (3.477)[4]	3.6702 (3.651)[7]
d^2	Mo	3.1998 (3.201)[1]	3.2685 (3.270)[1]	3.4970 (3.469)[2]
	W	3.1908 (3.222)[1]	3.2574	3.4970 (3.496)[3]
d^3	Tc	3.0692	3.1543	3.4149
	Re	3.0750	3.1311	3.3834
d^5	Co	Non-Layered Structure		3.5983 (3.802)[8]
	Rh			3.7563 (3.92)[8]
	Ir			3.8431 (3.928)[8]
d^6	Ni	3.3174	3.4712	3.7248 (3.854)[8]
	Pd	3.5408	3.6759	3.9162 (4.037)[8]
	Pt	3.5237 (3.543)[4]	3.6662 (3.728)[4]	3.9554 (4.026)[8]

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