Electronic Supplementary Material (ESI) for Materials Horizons.

Design of High Polarization Low Switching Barrier Hybrid Improper Ferroelectric Perovskite Oxide Superlattices

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Supporting information

High Polarization and Low Switching Barrier:

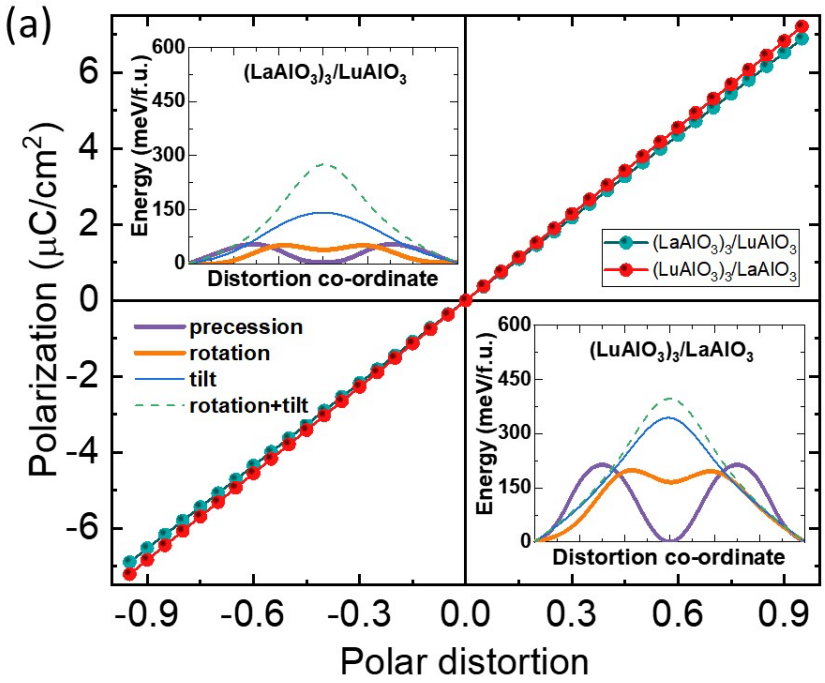

Figure S1. Comparison of Polarization in $(LaAlO_3)_1/(LuAlO_3)_3$ and $LuAlO_3)_1/(LaAlO_3)_3$ as a function of normalized polar distortion. The inset figures show the switching barrier for the out-of-phase rotation, precession and conventional rotation and conventional tilt switching path obtained from NEB calculations for the respective system

Energy barriers for different pathways:

Table S1: Energy barriers from NEB

A	A'	Rotation barrier (meV)	out-of-phase barrier (meV)	Rot+tilt barrier (Via H.S.) (meV)	Tilt barrier (meV)	Precession barrier (meV)
Ce	Dy	168.9	82.4	516.5	356.2	83.2
Ce	Gd	102.4	46.6	373.1	277.1	47.1
Ce	La	12.4	10.7	25.1	20.4	10.9
Ce	Lu	339.6	167.8	899.8	560.8	168.0
Ce	Nd	9.0	4.5	128.9	125.1	5.4

Ce	Sm	42.3	16.8	230.6	189.8	17.6
Ce	Tm	287.5	142.3	772.9	495.4	144.7
Ce	Y	218.5	108.4	511.8	296.5	116.2
Dy	Ce	41.4	19.1	191.4	154.9	20.1
Dy	Gd	190.3	91.6	549.6	368.1	92.0
Dy	La	43.8	21.7	181.0	142.5	22.0
Dy	Lu	440.6	220.1	1098.3	664.0	223.4
Dy	Nd	60.4	28.5	274.6	214.4	29.0
Dy	Sm	112.0	52.4	392.9	281.5	53.1
Dy	Tm	390.5	192.7	944.8	555.6	199.9
Dy	Y	268.5	130.8	713.3	451.2	140.0
Gd	Ce	25.2	12.5	151.0	131.3	13.3
Gd	Dy	241.5	120.1	651.6	413.8	124.9
Gd	La	27.3	9.7	139.6	115.8	10.1
Gd	Lu	420.2	209.6	1045.4	634.7	214.4
Gd	Nd	42.7	17.2	232.1	195.6	17.4
Gd	Sm	90.4	41.7	346.5	264.5	41.8
Gd	Tm	362.4	180.5	890.1	530.3	188.3
Gd	Y	241.6	120.4	660.2	423.2	122.4
La	Ce	1.6	1.0	49.9	48.6	1.9
La	Dy	167.8	83.3	513.1	345.7	83.9
La	Gd	102.6	50.3	369.8	276.9	50.4
La	Lu	345.7	209.4	420.8	369.0	237.2
La	Nd	8.5	5.7	123.4	121.0	5.9
La	Sm	42.2	17.7	226.8	185.2	18.3
La	Tm	276.7	134.2	743.6	470.5	140.8
La	Y	168.8	83.5	517.6	351.1	84.0
Lu	Ce	78.6	35.4	283.1	211.5	35.7
Lu	Dy	313.4	155.3	819.8	506.7	160.8
Lu	Gd	217.1	106.3	641.1	433.2	113.4
Lu	La	240.0	73.5	293.2	153.9	74.1
Lu	Nd	107.0	51.7	375.4	270.0	52.4
Lu	Sm	144.2	67.5	490.3	350.4	67.8
Lu	Tm	816.4	403.7	1197.0	384.3	409.3
Lu	Y	221.5	109.3	762.7	549.9	113.6
Nd	Ce	3.2	2.4	78.6	76.1	3.2
Nd	Dy	188.4	90.8	551.3	363.7	91.1
Nd	Gd	118.7	55.7	406.0	291.8	56.2
Nd	La	2.6	1.9	65.0	62.9	2.1
Nd	Lu	363.5	178.6	937.3	578.3	188.5
Nd	Sm	52.4	23.6	259.6	209.6	23.7
Nd	Tm	303.6	146.8	785.1	485.6	153.0
Nd	Y	189.5	93.1	558.0	372.5	93.2
Sm	Ce	10.9	5.9	109.9	99.2	6.8
Sm	Dy	212.5	102.4	596.4	386.9	109.3
Sm	Gd	139.6	67.2	447.9	316.8	67.9

Sm	La	11.7	6.2	97.3	86.4	6.5
Sm	Lu	385.8	189.1	985.9	606.0	198.5
Sm	Nd	17.8	4.5	184.4	176.3	4.9
Sm	Tm	330.8	164.1	832.2	508.7	167.0
Sm	Y	212.9	102.6	603.8	395.7	110.3
Tm	Ce	74.6	35.4	263.5	198.3	36.3
Tm	Dy	308.1	153.6	782.5	481.9	158.0
Tm	Gd	228.5	112.6	626.1	398.8	122.5
Tm	La	74.1	34.3	248.3	176.7	35.0
Tm	Lu	508.2	347.6	608.9	528.3	355.7
Tm	Nd	91.3	43.2	344.1	252.8	43.9
Tm	Sm	146.8	70.2	465.8	322.4	71.0
Tm	Y	308.5	153.6	792.9	492.1	163.4
Y	YCe	50.2	22.1	185.8	137.3	22.1
Y	YDy	267.9	132.3	706.3	442.1	141.4
Y	YGd	190.2	94.9	551.9	370.8	95.0
Y	YLa	44.7	21.7	179.8	141.8	22.2
Y	YLu	441.2	216.4	1102.0	663.1	223.5
Y	YNd	61.1	30.2	275.3	222.7	30.6
Y	YSm	112.4	52.9	394.3	284.5	53.2
Y	YTm	390.4	194.5	949.0	560.9	198.1

Finite temperature AIMD:

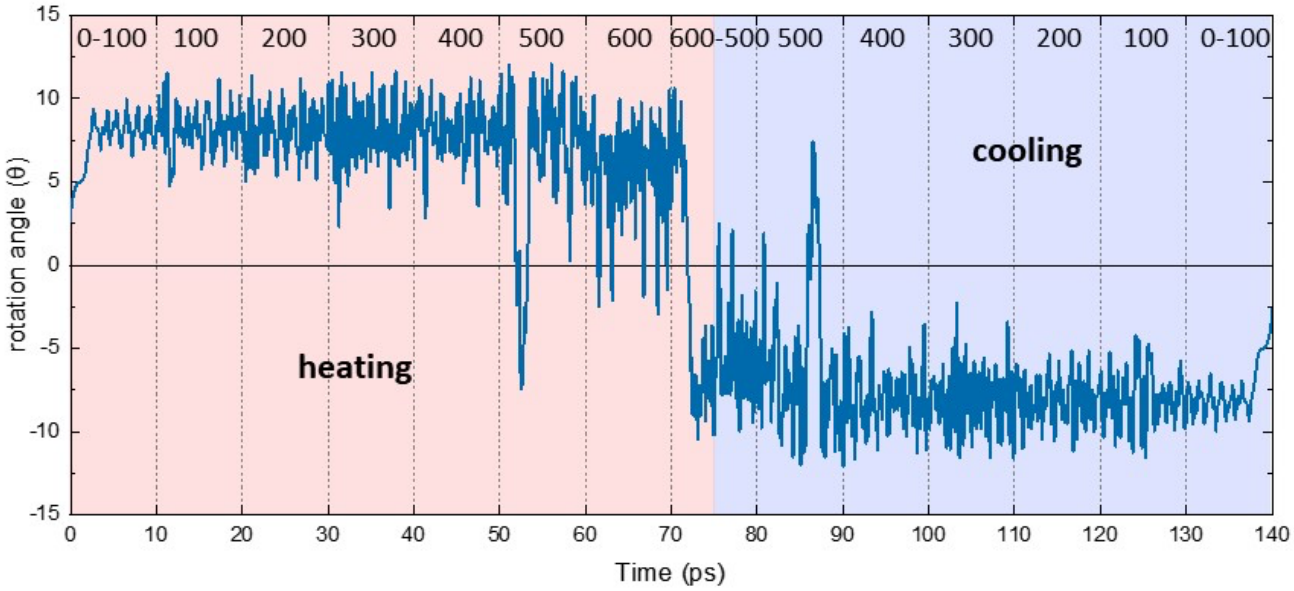

Figure S2 Average rotation angle θ_r as a function of temperature. Here the red and blue shaded areas indicate heating and cooling period. The system is heated till 600K and maintained until it switches its polarization. Then cooled immediately to maintain and prevent the back switching.

Polarization-Magnetization combined switching:

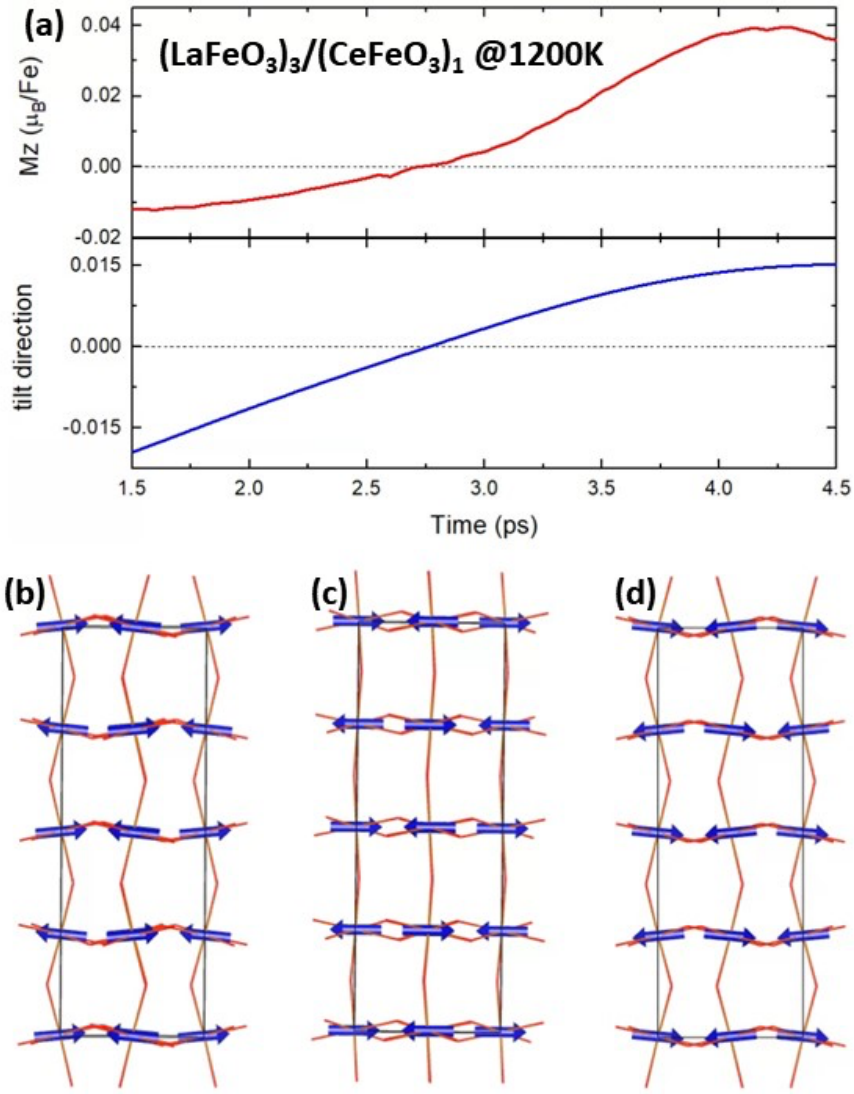

Figure S3 Polarization-Magnetization combined switching. AIMD simulation result of (LaFeO3)3/(CeFeO3)1. (a) Magnetization switching via tilt switching, the Mz and tilt direction as a function of simulation time. (b) to (d) Schematic showing the switching of Mz.