Phonon calculation

The phonon band structures for $LaSrMnO_4$ (LSMO) and $LaSrTiO_4$ (LSTO) were calculated using the PBE functional [1] and the projected augmented wave method [2] as implemented in the VASP code. The in-plane a (b) and out-of-plane c lattices of the three cationic arrangements AABB, ABAB and ABBA were relaxed till the forces less than 0.1 meV/Å with a convergence of the ionic steps less than 10^{-8} eV. An energy cutoff of 550 eV and Γ-centred k-point mesh of 8×8×5 were used. The phonon band structures of these configurations were calculated using a 2×2×2 supercell in the finite difference method using the PHONOPY code [3]. For the ABBA configuration, the primitive cell was also relaxed using the PBEsol functional [4] which results in all positive frequencies while the PBE functional shows doubly negative frequency at the M (0.5, 0.5,0) and A (0.5, 0.5, 0.5). Since PBE is known to the overestimate the lattice constants which can significantly affect the phonon frequencies [5], the results from PBEsol are presented. The phonon band structures for LSMO and LSTO within different layer stackings are shown in Fig. S1. The optimized lattices and the magnetic and electronic properties within the PBE method are also summarized in Table S1.

Table S1. Fully relaxed in-plane lattice a and out-of-plane lattice c

		AABB	ABAB	ABBA[a]
$LaSrMnO_4$	a (Å)	3.80	3.80	3.80
	c (Å)	12.72	12.59	12.58
$LaSrTiO_4$	a (Å)	3.94	3.93	3.88
	c (Å)	12.67	12.64	12.51

[a]The results are obtained from PBEsol functional

[1] J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. 77, 3865 (1996).

[2] G. Kresse and D. Joubert, Phys. Rev. B 59, 1758 (1999).

[3] A. Togo and I. Tanaka, *Scr. Mater.* **108**, 1-5 (2015).

[4] J.P. Perdew, A. Ruzsinszky, G.I. Csonka, O.A. Vydrov, G.E. Scuseria, L.A. Constantin, X. Zhou, and K. Burke, *Phys. Rev. Lett.* **100,** 136406 (2008).

[5] G. I. Csonka, J. P. Perdew, A. Ruzsinszky, P. H. T. Philipsen, S. Lebègue, J. Paier, O. A. Vydrov, and J. G. Ángyán, *Phys. Rev. B* **79**, 155107 (2009).

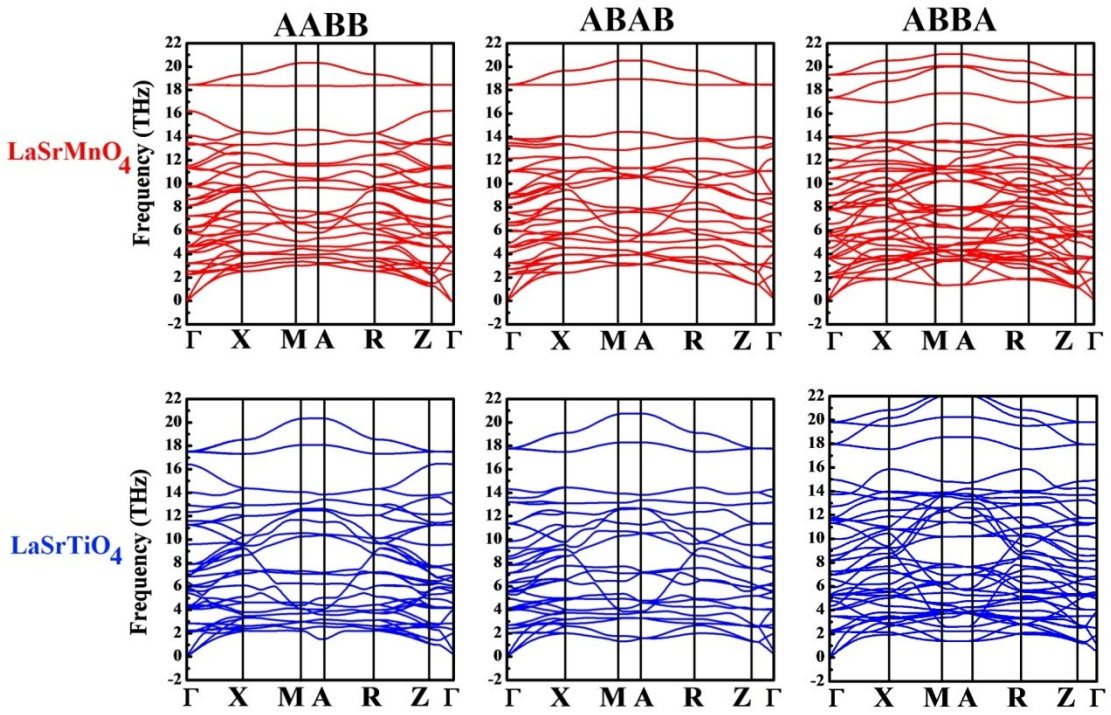

FIG. S1. The phonon band structure of the Ruddlesden-Popper $LaSrMnO_4$ and $LaSrTiO_4$ in different stacking configurations: AABB, ABAB and ABBA.

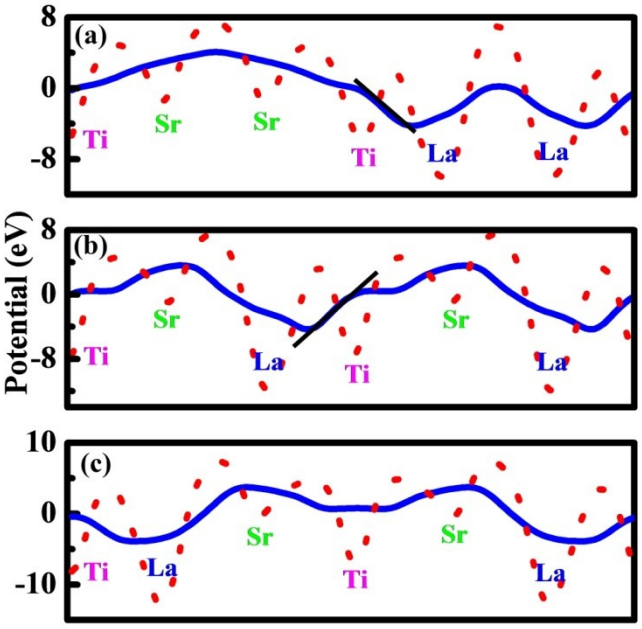

FIG. S2. The local potentials and their averages calculated for different configurations using the HSE06 functional: a) AABB, b) ABAB, and c) ABBA. The dotted lines represent the total potentials containing only the electrostatic contribution (ionic plus Hartree), while the blue thick lines are the averages of the potential along the [001] direction. The averaging is performed over a window of 2.01 Å which corresponds to the inter-layer distance. The linear lines in (a) and (b) show the slopes which were used to calculate the internal electric fields in configuration AABB and ABAB.