Predicting the Structural, Electronic and Magnetic Properties of Few Atomic-layer Polar Perovskite

Shaowen Xu[a,b*], Fanhao Jia[a,b*], Shunbo Hu[b,a], Athinarayanan Sundaresan[c], Nikita V.Ter-Oganessian[d] Alexander P. Pyatakov[e], Jinrong Cheng[b,a], Jincang Zhang[b,a], Shixun Cao[a,b], and Wei Ren[a,b,†]

[a]*Physics Department, International Center for Quantum and Molecular Structures, Shanghai Key Laboratory of High Temperature Superconductors, Shanghai University, Shanghai 200444, China*

[b]*Materials Genome Institute, State Key Laboratory of Advanced Special Steel, School of Materials Science and Engineering, Shanghai University, Shanghai 200444, China*

[c]*Chemistry and Physics of Materials Unit, Jawaharlal Nehru Centre for Advanced Scientific Research, Bangalore-560064, India*

[d]*Institute of Physics, Southern Federal University, Rostov-on-Don, 344090, Russia*

[e]*M.V. Lomonosov Moscow State University, Faculty of Physics 1-2 Leninskiye Gory, GSP-1, Moscow, 119991, Russia*

[†]renwei@shu.edu.cn

*These authors contributed equally to this work.

Table S1. The elastic constants (N/m) of few-AL KTaO systems. The criteria of elastic stability in 2D systems were described in Ref [1], where a stable structure should simultaneously satisfy $C_{11} > 0$ & $C_{66} > 0$ & $C_{11} > |C_{12}|$ for 2AL and Type I 3AL ($3AL_I$) cases, while the Type II 3AL ($3AL_{II}$) system should meet $C_{11} > 0$ &$C_{66} > 0$ & $C_{11}C_{22} > C_{12}C_{12}$.

System	Elastic Constants (N/m)			
	C_{11}	C_{12}	C_{66}	C_{22}
2AL	124.8	35.3	6.4	/
$3AL_I$	135.0	46.2	18	/
$3AL_{II}$	168.7	45.8	8.0	45.8

Table S2. The PBE optimized in-plane lattice parameter *a* (Å) which is defined by the distance between adjacent K atom. The energy gap (eV) was calculated by using the HSE06 method.

System	a (Å)	Gap (eV)
2AL	3.919	3.51
4AL	3.941	3.21
6AL	3.981	3.10
8AL	3.990	2.75
10AL	3.997	2.17
$Bulk_{cubic}$	3.984	3.64[2]

Table S3. The PBE optimized in-plane lattice parameter a (Å) which is defined by the distance between adjacent K atom. The magnetic moment M (μ_B/u.c.) of odd-AL systems, where the "*u.c.*" is defined by the in-plane area of EL unit-cell displayed in Fig. 1.

System	a (Å)		M (μ_B/uc)	
	Type I	Type II	Type I	Type II
3AL	4.038	3.922	1	0
5AL	4.039	3.989	0.93	0.92
7AL	4.042	3.997	0.76	0.75
9AL	4.043	4.004	0.48	0.28

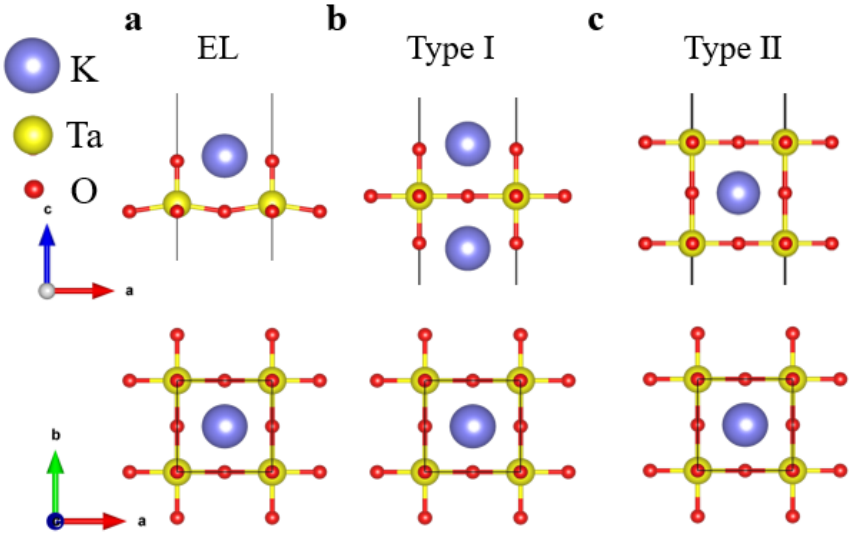

Figure S1. The side and top views of the ideal structures of 2D (**a)** $KTaO_3$ (EL), (**b)** K_2TaO_4 (Type I), and (**c)** KTa_2O_5 (Type II).

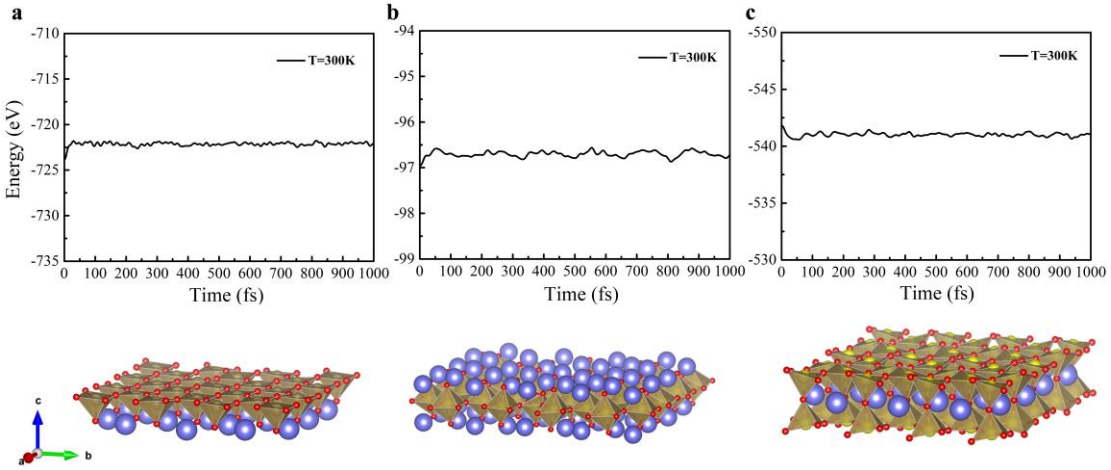

Figure S2. The *ab* initio molecular dynamics (AIMD) simulations were carried out for 1 *ps* with a time step of 1 *fs* at room temperature (300 K) within the canonical ensemble.

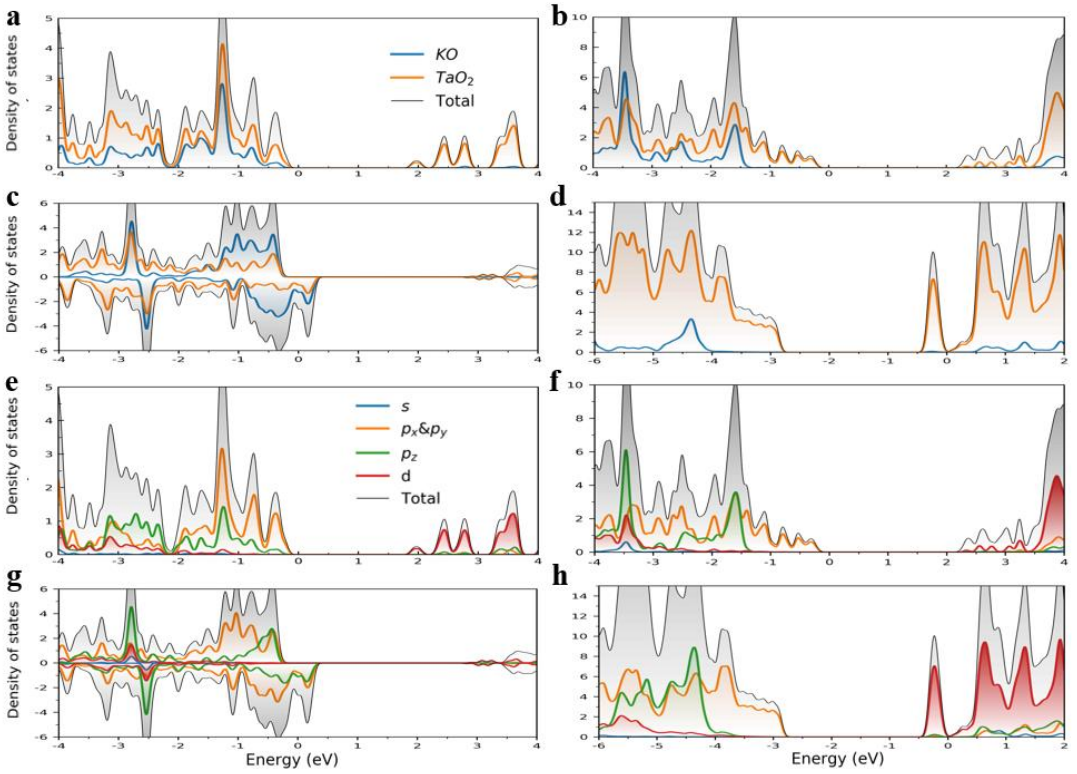

Figure S3. (a)-(d) are the layer resolved projected density of states (PDOS). (e)-(h) are the atomic orbital PDOS. (a) and (e) are for bulk KTO; (b) and (f) are for EL (2AL) system; (c) and (g) are for Type I (3AL); (d) and (h) are for Type II (3AL).

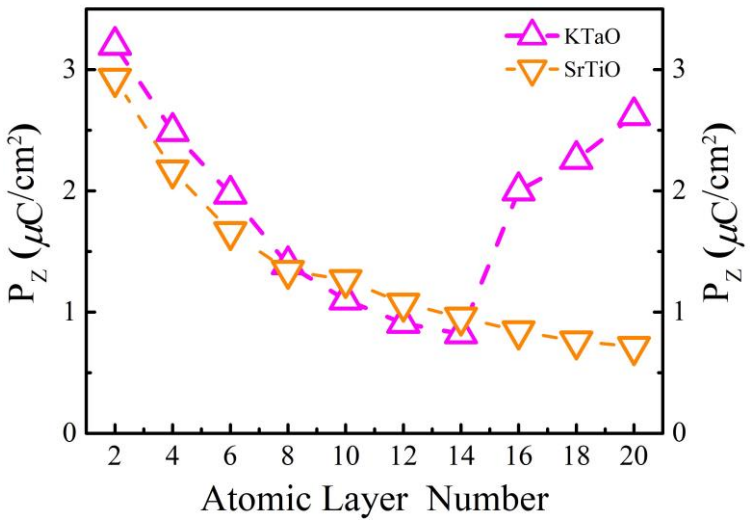

Figure S4. The out-of-plane polarization P_z (μC/cm^2) of EL KTaO systems and EL SrTiO systems as functions of AL thickness. The P_z was calculated by integrating the charge density, which is dependent on the effective thickness of slab. Here, we applied an additional 2.5 Å space on each side of the slab to calculate the effective volume Ω.

Reference

[1] M. Maździarz, 2D Materials **6**, 048001 (2019).

[2] G. Jellison Jr, I. Paulauskas, L. Boatner and D. Singh, Physical Review B **74**, 155130 (2006).