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

Supporting Information

First-Principles Studies of Polar Perovskite $KTaO_3$ Surfaces: Structural Reconstruction, Charge Compensation, and Stability Diagram

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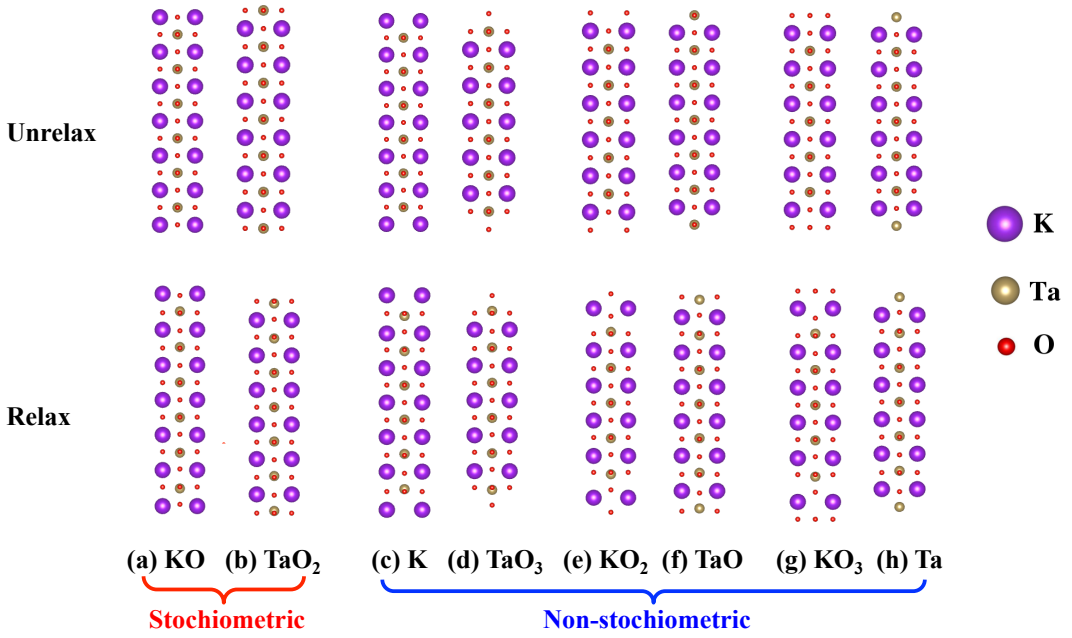

FIG. S1: (Color online) Unrelaxed and relaxed slab models of $KTaO_3$ (001) surface with various terminations: (a) KO, (b) TaO_2, (c) K, (d) TaO_3, (e) KO_2, (f) TaO, (g) KO_3, and (h) Ta.

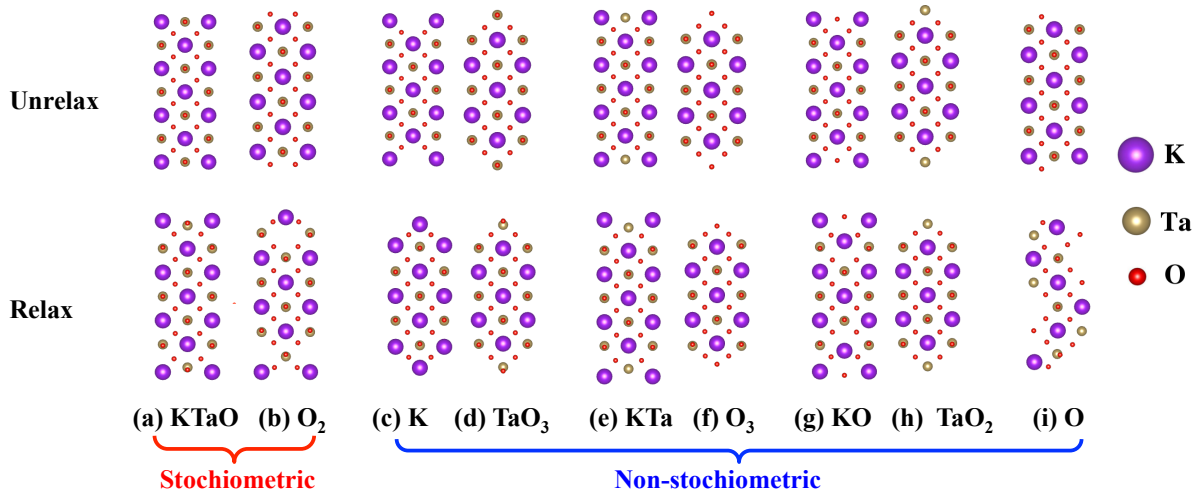

FIG. S2: (Color online) Unrelaxed and relaxed slab models of $KTaO_3$ (110) surface with various terminations: (a) KTaO, (b) O_2, (c) K, (d) TaO_3, (e) KTa, (f) O_3, (g) KO, (h) TaO_2, and (i) O. Note that the surface model with O termination (i) is symmetrical to its complementary surface model and thus only one surface is shown.

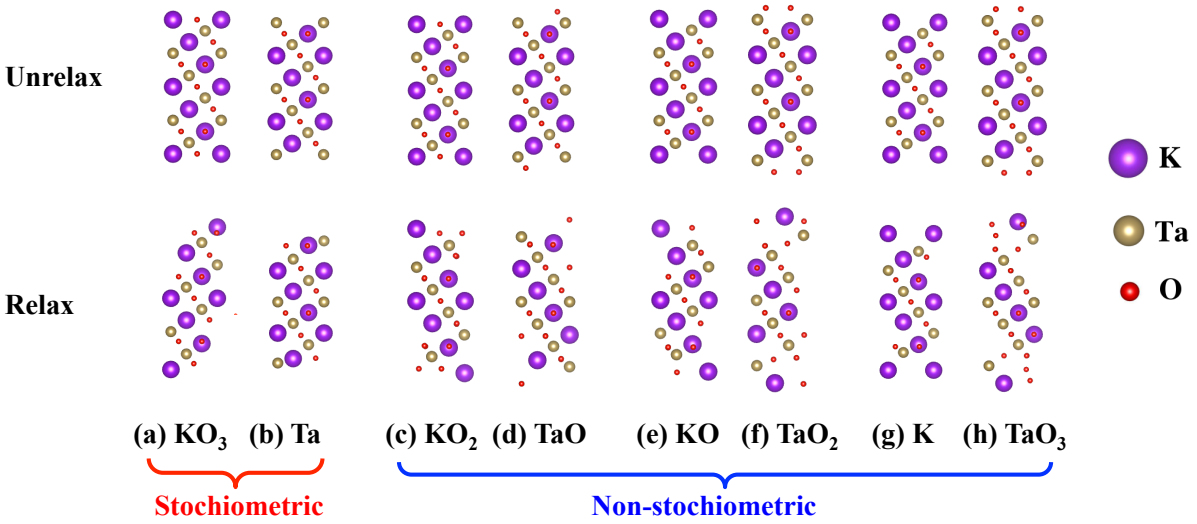

FIG. S3: (Color online) Unrelaxed and relaxed slab models of $KTaO_3$ (111) surface with various terminations: (a) KO_3, (b) Ta, (c) KO_2, (d) TaO, (e) KO, (f) TaO_2, (g) K, and (h) TaO_3.

TABLE S1: Calculated Bader charge (in e) for the (001) surface of $KTaO_3$. $\sum_{l=1}^{6} \sigma$ and σ_7 are the sum of the charge for the six outermost layers and the seventh layer, respectively.

Terminations		Stochimetric		Non-stochiometric					
		KO	TaO_2	K	TaO_3	KO_2	TaO	KO_3	Ta
L1	K	0.85		0.58					0.39
	Ta		2.66				1.54		
	O	-1.21	-1.19		-0.58	-0.35	-1.22	-0.21(-0.18)	
L2	K		0.82			0.87	0.80	0.87	0.76
	Ta	2.83		2.24	2.66				
	O	-1.21	-1.10	-1.26	-1.05	-0.91	-1.04	-0.95	-1.08
L3	K	0.83		0.81	0.83				
	Ta		2.80			2.68	2.72	2.69	2.69
	O	-1.20	-1.23	-1.08	-1.05	-1.15	-1.23	-1.12	-1.22
L4	K		0.81			0.83	0.81	0.82	0.81
	Ta	2.79		2.78	2.67				
	O	-1.18	-1.20	-1.23	-1.18	-1.14	-1.18	-1.12	-1.16
L5	K	0.81		0.80	0.81				
	Ta		2.78			2.77	2.77	2.77	2.76
	O	-1.13	-1.22	-1.22	-1.13	-1.17	-1.22	-1.17	-1.22
L6	K		0.80			0.80	0.80	0.80	0.80
	Ta	2.78		2.75	2.79				
	O	-1.19	-1.23	-1.22	-1.19	-1.13	-1.24	-1.13	-1.24
L7	K	0.81		0.80	0.81				
	Ta		2.76			2.79	2.74	2.79	2.74
	O	-1.14	-1.22	-1.25	-1.13	-1.18	-1.22	-1.19	-1.22
$\sum_{l=1}^{6} \sigma$		0.17	-0.16	0.23	0.17	-0.20	-0.15	-0.20	-0.14
σ_7		-0.33	0.33	-0.45	-0.32	0.42	0.31	0.41	0.30

TABLE S2: Calculated Bader charge (in e) for the (110) surface of $KTaO_3$. $\sum_{l=1}^{6}\sigma$ and σ_7 are the sum of the charge for the six outermost layers and the seventh layer, respectively. The number in the parentheses is for O ions in the same layer but with different Bader charges.

Terminations		Stochimetric		Non-stochiometric						
		KTaO	O_2	K	TaO_3	KTa	O_3	KO	TaO_2	O
L1	K	0.78		0.83		0.71		0.88		
	Ta	1.98			2.45				1.27	
	O	-1.27	-0.92		-1.23	0.79	-0.09	-0.51		-1.11
L2	K		0.88							0.86
	Ta		2.73							2.64
	O	-1.17	-1.07	-1.08	-1.10	-1.17	-0.68(-0.69)	-0.96	-1.10	-1.24
L3	K	0.80		0.83	0.81	0.79	0.81	0.84	0.80	
	Ta	2.69		2.69	2.73	2.69	2.71	2.72	2.71	
	O	-1.23	-1.08	-1.22	-1.19	-1.22	-1.08	-1.13	-1.20	-1.20(-1.15)
L4	K		0.81							0.82
	Ta		2.79							2.84
	O	-1.22	-1.13	-1.16	-1.21	-1.21	-1.13	-1.11	-1.21	-1.21
L5	K	0.81		0.82	0.81	0.81	0.82	0.81	0.81	
	Ta	2.69		2.79	2.70	2.67	2.76	2.84	2.69	
	O	-1.22	-1.13	-1.17	-1.22	-1.22	-1.18	-1.15	-1.22	-1.21(-1.19)
L6	K		0.81							0.81
	Ta		2.74							2.76
	O	-1.17	-1.17	-1.18	-1.27	-1.16	-1.18	-1.20	-1.17	-1.21
L7	K	0.81		0.81	0.82	0.81	0.81	0.81	0.81	
	Ta	2.60		2.87	2.63	2.57	2.87	2.87	2.59	
	O	-1.23	-1.14	-1.18	-1.23	-1.24	-1.18	-1.17	-1.23	-1.22(-1.20)
$\sum_{l=1}^{6}\sigma$		-1.07	1.14	-1.24	- 1.09	-1.06	-1.24	-1.25	-1.07	1.21
σ_7		2.17	-2.28	2.5	2.22	2.14	2.50	2.50	2.17	-2.41

TABLE S3: Calculated Bader charge (in e) for the (111) surface of $KTaO_3$. $\sum_{l=1}^{6}\sigma$ and σ_7 are the sum of the charge for the six outermost layers and the seventh layer, respectively. The number in the parentheses is for O ions in the same layer but with different Bader charges.

Terminations		Stochimetric		Non-stochiometric					
		KO_3	Ta	KO_2	TaO	KO	TaO_2	K	TaO_3
L1	K	0.89		0.88		0.83		0.73	
	Ta		1.72		2.48		2.69		2.72
	O	-0.87(-0.86)		-1.06(-1.05)	-0.89	-1.25	-0.80(-0.83)		-0.59(-0.80/-0.76)
L2	K		0.82		0.84		0.90		0.90
	Ta	2.70		2.62		2.48		1.24	
	O		-1.17		-1.21(-1.22)		-1.16		-0.68(-1.14)
L3	K	0.82		0.82		0.78		0.78	
	Ta		2.68		2.80		2.86		2.82
	O	-1.13		-1.21(-1.18)		-1.25(-1.23)		-1.21	
L4	K		0.78		0.81		0.81		0.82
	Ta	2.79		2.80		2.63		2.60	
	O		-1.21		-1.21(-1.22)		-1.18(-1.19)		-1.19(-1.17)
L5	K	0.82		0.82		0.81		0.8	
	Ta		2.67		2.73		2.82		2.82
	O	-1.16		-1.21		-1.22		-1.22(-1.21)	
L6	K		0.82		0.82		0.82		0.82
	Ta	2.80		2.81		2.56		2.56	
	O		-1.22		-1.21		-1.21		-1.20(-1.21)
L7	K	0.82		0.82		0.81		0.81	
	Ta		2.63		2.70		2.80		2.80
	O	-1.18		-1.21		-1.22		-1.22(-1.21)	
$\sum_{l=1}^{6}\sigma$		1.36	-1.30	1.42	-1.34	1.44	-1.39	1.44	-1.39
σ_7		-2.71	2.63	-2.83	2.70	-2.84	2.80	-2.84	2.80