

Supporting information

Photophysical properties of SrTaO₂N thin films and impact of the anion ordering: a joined theoretical and experimental investigation

Ahmed Ziani^a, Claire Le Paven^b, Laurent Le Gendre^b, Florent Marlec^b, Ratiba Benzerga^b, Franck Tessier^c, François Cheviré^c, Mohamed N. Hedhili^d, Angel T. Garcia-Esparza^e, Sigismund Melissen^e, Philippe Sautet^{e,f}, Tanguy Le Bahers^e, Kazuhiro Takanabe^{a*}

^aKing Abdullah University of Science and Technology (KAUST), KAUST Catalysis Center (KCC) and Physical Sciences and Engineering Division (PSE), 4700 KAUST, Thuwal, 23955-6900, Saudi Arabia.

^bInstitut d'Electronique et de Télécommunications de Rennes (IETR), Equipe FunMAT, IUT Saint-Brieuc, Université de Rennes 1, 22000 Saint Brieuc, France.

^cInstitut des Sciences Chimiques de Rennes (ISCR), Equipe Verres et Céramiques, Université de Rennes 1, 35042 Rennes cedex, France.

^dKing Abdullah University of Science and Technology (KAUST), Imaging and Characterization Core Lab, 4700 KAUST, Thuwal, 23955-6900, Saudi Arabia

^eUniv Lyon, ENS de Lyon, CNRS, Université Claude Bernard Lyon 1, Laboratoire de Chimie UMR 5182, F-69342, Lyon, France

^fDepartment of Chemical and Biomolecular Engineering, University of California Los Angeles (UCLA), Los Angeles, California 90095, United States

** Email for corresponding author: kazuhiro.takanabe@kaust.edu.sa*

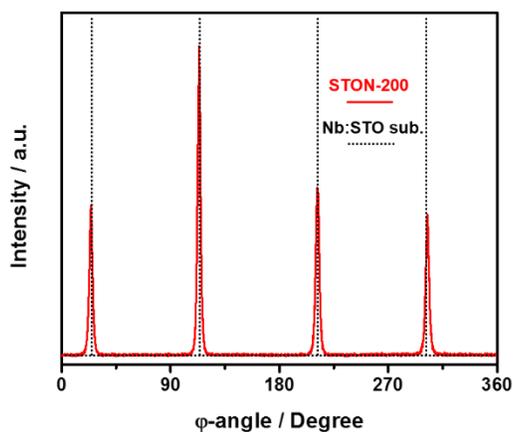


Figure S1. X-ray diffraction φ -scans at $\chi = 45^\circ$ of the STON-200 film and Nb:STO substrate associated with the (110) plane of the pseudo-cubic cells.

Table S1. The chemical composition of samples was determined by semi-quantitative Energy Dispersive Spectrometry (EDS).

Sample	O (% at.)	N (% at.)	Sr (% at.)	Ta (% at.)	Sr/Ta ratio
STON-200	36.86	13.01	25.77	23.74	1.085
STON-400	35.06	13.44	24.52	22.65	1.083
STON-900	35.60	13.22	25.78	23.86	1.081

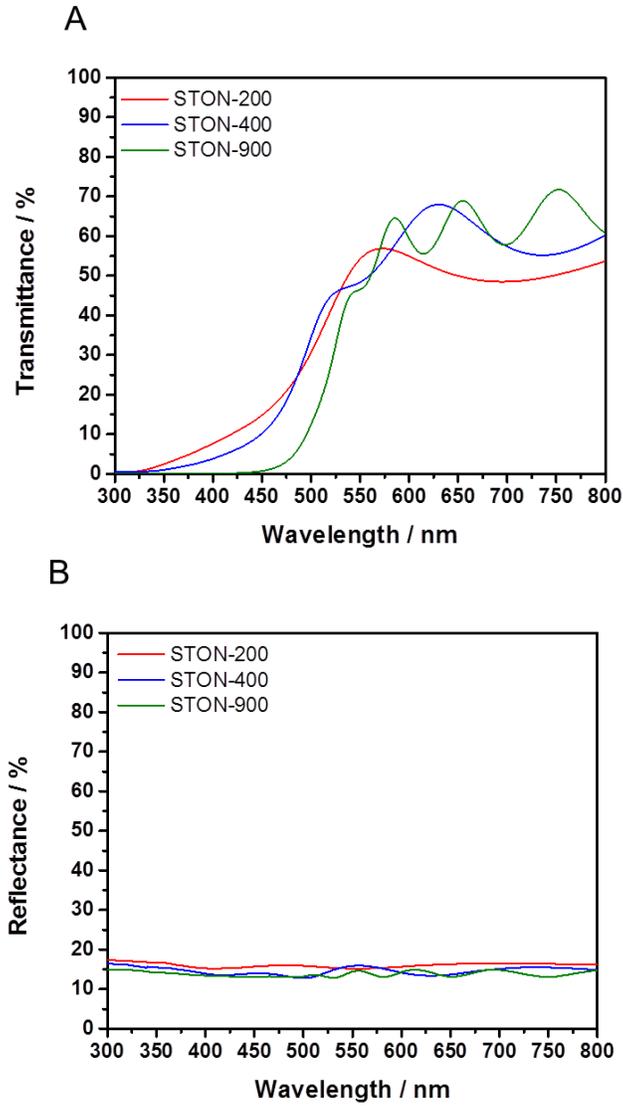


Figure S2. Transmittance (A) and reflectance (B) spectra of the measured SrTaO₂N films.

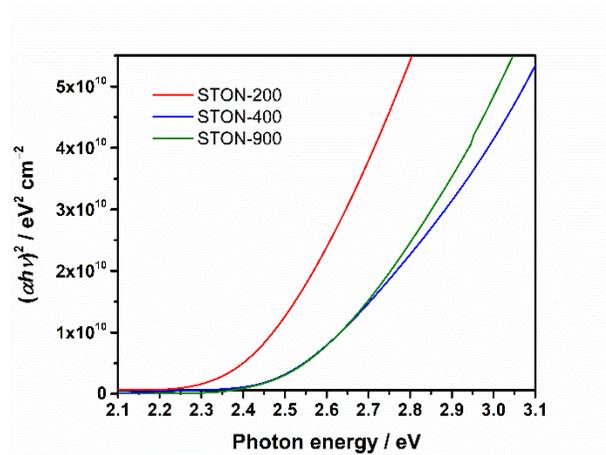


Figure S3. Tauc plots for direct allowed transition of the SrTaO₂N films.

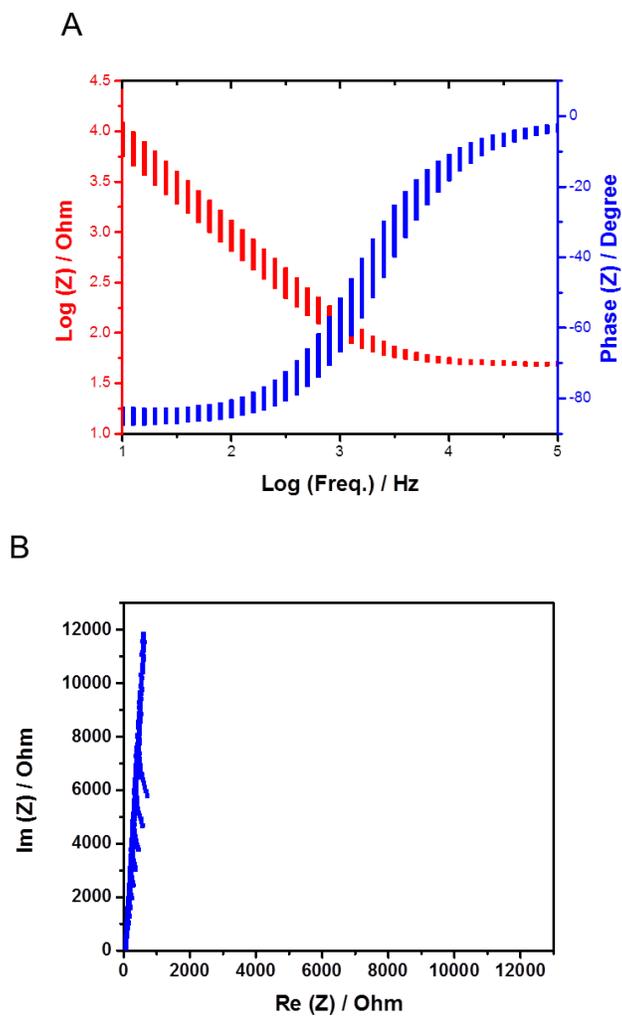


Figure S4. Bode (A) and Nyquist impedance spectra (B) of the measured STON-200 film.

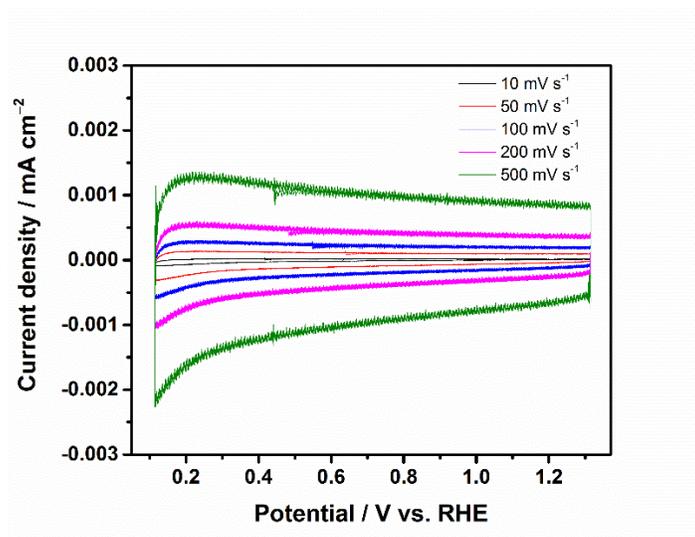


Figure S5. CV measurements of the measured STON-200 film with different scan rates.

Table S2. Experimental and computed cell parameters for different anion ordering in the $I4/mcm$ space group.

	STON-200	STON-400	STON-900	S1	S2	S3	S4	S5	S6
a / Å	5.730	5.731	5.736	5.728	5.645	5.644	5.727	5.716	5.651
c / Å	8.133	8.125	8.154	7.889	8.107	8.110	7.913	7.909	8.101

Table S3. Geometry of structure (S1)

a, b, c / Å	5.728	5.715	7.889
$\alpha, \beta, \gamma / ^\circ$	90.00	90.00	89.97
Fractional Coordinates			
Ta	0.4837	0.4894	-0.4992
Ta	0.0164	-0.0106	0.0012
Ta	0.4837	0.4893	-0.0011
Ta	0.0164	-0.0105	0.4990
Sr	-0.0155	0.4945	0.2501
Sr	-0.4838	-0.0056	-0.2499
Sr	-0.4863	-0.0059	0.2501
Sr	-0.0137	0.4943	-0.2500
N	0.2511	0.2520	-0.0187
N	0.2488	-0.2481	0.0200
N	0.2508	0.2522	-0.4815
N	0.2491	-0.2478	0.4800
O	-0.2473	0.2599	-0.4787
O	-0.2527	-0.2402	0.4800
O	-0.2469	0.2605	-0.0212
O	-0.2533	-0.2395	0.0198
O	-0.0217	0.0438	0.2500
O	-0.0200	-0.0357	-0.2500
O	-0.4813	-0.4562	-0.2500
O	-0.4776	0.4642	0.2500

Table S4. Geometry of structure (S2)

a, b, c / Å	5.645	5.645	8.107
$\alpha, \beta, \gamma / ^\circ$	90.05	90.09	90.02
Fractional Coordinates			
Ta	-0.5000	-0.4994	0.4854
Ta	0.0039	0.0012	-0.0142
Ta	0.4926	0.4865	0.0018
Ta	0.0138	-0.0122	-0.4988
Sr	0.0006	0.4931	0.2456
Sr	-0.4938	-0.0085	-0.2525
Sr	-0.4915	-0.0043	0.2465
Sr	0.0001	0.4949	-0.2553
N	0.2770	0.2262	0.0024
N	-0.4851	0.4928	0.2518
N	0.2758	-0.2220	0.4943
N	0.0006	-0.0072	-0.2481
O	-0.2182	0.2864	-0.0076
O	0.2195	-0.2764	0.0133
O	-0.2868	0.2203	-0.4843
O	-0.2817	-0.2123	0.0063
O	0.2153	0.2829	-0.4968
O	-0.2204	-0.2805	-0.4989
O	-0.0233	0.0191	0.2548
O	-0.4984	-0.4805	-0.2456

Table S5. Geometry of structure (S3)

a, b, c / Å	5.644	5.639	8.110
$\alpha, \beta, \gamma / ^\circ$	90.00	90.00	90.81
Fractional Coordinates			
Ta	0.4972	-0.4987	0.4857
Ta	0.0029	-0.0011	-0.0141
Ta	0.4861	-0.4889	0.0020
Ta	0.0139	-0.0109	-0.4978
Sr	-0.0048	0.4959	0.2477
Sr	-0.4952	0.0048	-0.2523
Sr	-0.4918	0.0037	0.2459
Sr	-0.0082	0.4964	-0.2542
N	-0.4938	0.4932	0.2517
N	0.2771	-0.2199	-0.4972
N	-0.0067	0.0073	-0.2482
N	0.2228	-0.2803	0.0022
O	0.2877	0.2148	0.0031
O	-0.2109	0.2829	0.0045
O	-0.2894	0.2167	-0.4960
O	-0.2789	-0.2185	0.0026
O	0.2123	0.2851	-0.4968
O	-0.2210	-0.2817	-0.4975
O	-0.0120	0.0113	0.2544
O	-0.4872	0.4880	-0.2456

Table S6. Geometry of structure (S4)

a, b, c / Å	5.727	5.722	7.913
$\alpha, \beta, \gamma / ^\circ$	90.21	89.99	89.88
Fractional Coordinates			
Ta	-0.4849	0.4838	-0.4977
Ta	0.0154	-0.0161	0.0023
Ta	0.4847	0.4837	0.0018
Ta	-0.0150	-0.0163	-0.4982
Sr	-0.0005	0.4864	0.2532
Sr	-0.4998	-0.0145	-0.2464
Sr	-0.4983	-0.0132	0.2528
Sr	0.0010	0.4860	-0.2472
N	0.2576	0.2473	0.0166
N	0.2423	-0.2526	0.0221
N	-0.2590	0.2465	-0.4779
N	-0.2413	-0.2535	-0.4835
O	-0.2416	0.2704	-0.0265
O	-0.2592	-0.2292	-0.0204
O	0.2393	0.2727	0.4795
O	-0.0435	0.0176	0.2490
O	0.4565	-0.4822	-0.2510
O	-0.4568	-0.4947	0.2490
O	0.2598	-0.2277	0.4736
O	0.0433	0.0057	-0.2509

Table S7. Geometry of structure (S5)

a, b, c / Å	5.716	5.714	7.909
$\alpha, \beta, \gamma / ^\circ$	90.06	89.96	89.65
Fractional Coordinates			
Ta	-0.4886	0.4822	-0.4974
Ta	0.0173	-0.0122	0.0028
Ta	0.4870	0.4882	0.0010
Ta	0.0109	0.0129	-0.4997
Sr	-0.0016	0.4953	0.2513
Sr	-0.4885	-0.0085	-0.2453
Sr	-0.4932	-0.0154	0.2553
Sr	0.0033	-0.4966	-0.2488
N	0.2546	0.2501	0.0052
N	0.2455	-0.2497	0.0281
N	0.2498	0.2459	0.4936
N	-0.2505	0.2545	-0.4723
O	0.2334	-0.2563	0.4659
O	-0.2653	-0.2412	-0.4945
O	-0.2434	0.2670	-0.0341
O	-0.2580	-0.2344	-0.0074
O	-0.0474	0.0224	0.2492
O	0.0253	-0.0323	-0.2512
O	0.4662	-0.4627	-0.2507
O	-0.4568	0.4907	0.2490

Table S8. Geometry of structure (S6)

a, b, c / Å	5.651	5.664	8.101
$\alpha, \beta, \gamma / ^\circ$	89.99	90.15	88.77
Fractional Coordinates			
Ta	-0.4980	0.4977	0.4847
Ta	0.0020	-0.0021	-0.0153
Ta	0.4871	0.4821	0.0001
Ta	-0.0128	-0.0182	-0.4999
Sr	-0.0023	0.4889	0.2435
Sr	0.4975	-0.0114	-0.2563
Sr	0.4975	-0.0108	0.2439
Sr	-0.0030	0.4891	-0.2563
N	0.2755	0.2300	-0.0015
N	-0.4844	0.4814	0.2526
N	0.0157	-0.0187	-0.2474
N	-0.2243	-0.2703	0.4984
O	0.2800	-0.2176	0.4853
O	-0.2203	0.2822	-0.0147
O	0.2284	-0.2691	0.0248
O	-0.2719	0.2305	-0.4753
O	-0.2722	-0.2160	0.0110
O	0.2275	0.2842	-0.4890
O	-0.0110	0.0340	0.2556
O	0.4891	-0.4658	-0.2444

Table S9. Computed properties for each structure.

	S1	S2	S3	S4	S5	S6
E_g / eV	2.2	2.6	2.3	2.4	2.3	2.3
ϵ_∞	5.7	5.7	5.7	5.6	5.8	5.6
ϵ_{vib}	56.1	59.2	57.4	47.8	59.7	47.1
ϵ_r	61.8	64.9	63.1	53.4	65.5	52.7
m_e^* / m_e	0.5-0.5	0.3-0.4	0.6-0.8	0.5-0.7	0.4-0.8	0.4-0.6
m_h^* / m_e	0.7-1.7	0.8-1.6	0.6-1.1	0.7-2.0	1.3-4.8	0.4-0.9
E_b / meV	1.0	0.7	1.0	1.1	1.3	0.8