Table S1 – occupancies, starting bulk XYZ positions and final fit positions for top six atomic layers in room temperature best fit model. The X,Y,Z co-ordinates correspond to fractional positions along the a,b,c lattice directions.

El	Bulk X	Bulk Y	Bulk Z	Fit X	Error X	Fit Y	Error Y	Fit Z	Error Z	occval
O	0.0	0.0	1.0000	0.0906	0.027	-0.0822	0.075	1.0403	0.005	0.11
La/Sr	0.5	0.5	0.9783	0.4462	0.002	0.4464	0.002	0.9845	0.001	0.11
Al	0.0	0.0	0.8370	0.0126	0.007	-0.0161	0.010	0.8131	0.008	0.23
O	0.0	0.5	0.8370	0.1000	0.057	0.5963	0.046	0.8619	0.010	0.23
O	0.5	0.0	0.8370	0.4804	0.008	0.0950	0.005	0.8625	0.006	0.23
La/Sr	0.5	0.5	0.6957	0.5079	0.004	0.4919	0.001	0.7104	0.000	0.31
O	0.0	0.0	0.6740	-0.0370	0.017	0.0268	0.006	0.6755	0.002	0.31
O	0.5	0.5	0.5000	0.4797	0.003	0.5442	0.001	0.4997	0.002	0.85
La/Sr	0.0	0.0	0.4783	-0.0050	0.001	0.0098	0.001	0.4788	0.000	0.85
Al	0.5	0.5	0.3370	0.4462	0.005	0.5266	0.002	0.3327	0.001	0.95
O	0.5	0.0	0.3370	0.4729	0.006	0.0001	0.003	0.3325	0.000	0.95
O	0.0	0.5	0.3370	-0.0093	0.007	0.4540	0.002	0.3320	0.000	0.95
La/Sr	0.0	0.0	0.1957	0.0006	0.000	0.0052	0.000	0.1972	0.000	1.00
O	0.5	0.5	0.1740	0.5024	0.006	0.5018	0.001	0.1811	0.001	1.0

Figure S1 – Comparison of CTR structure factor data (black error lines), best-fits (red lines) and bulk profiles (dotted grey lines) from full room temperature CTR dataset

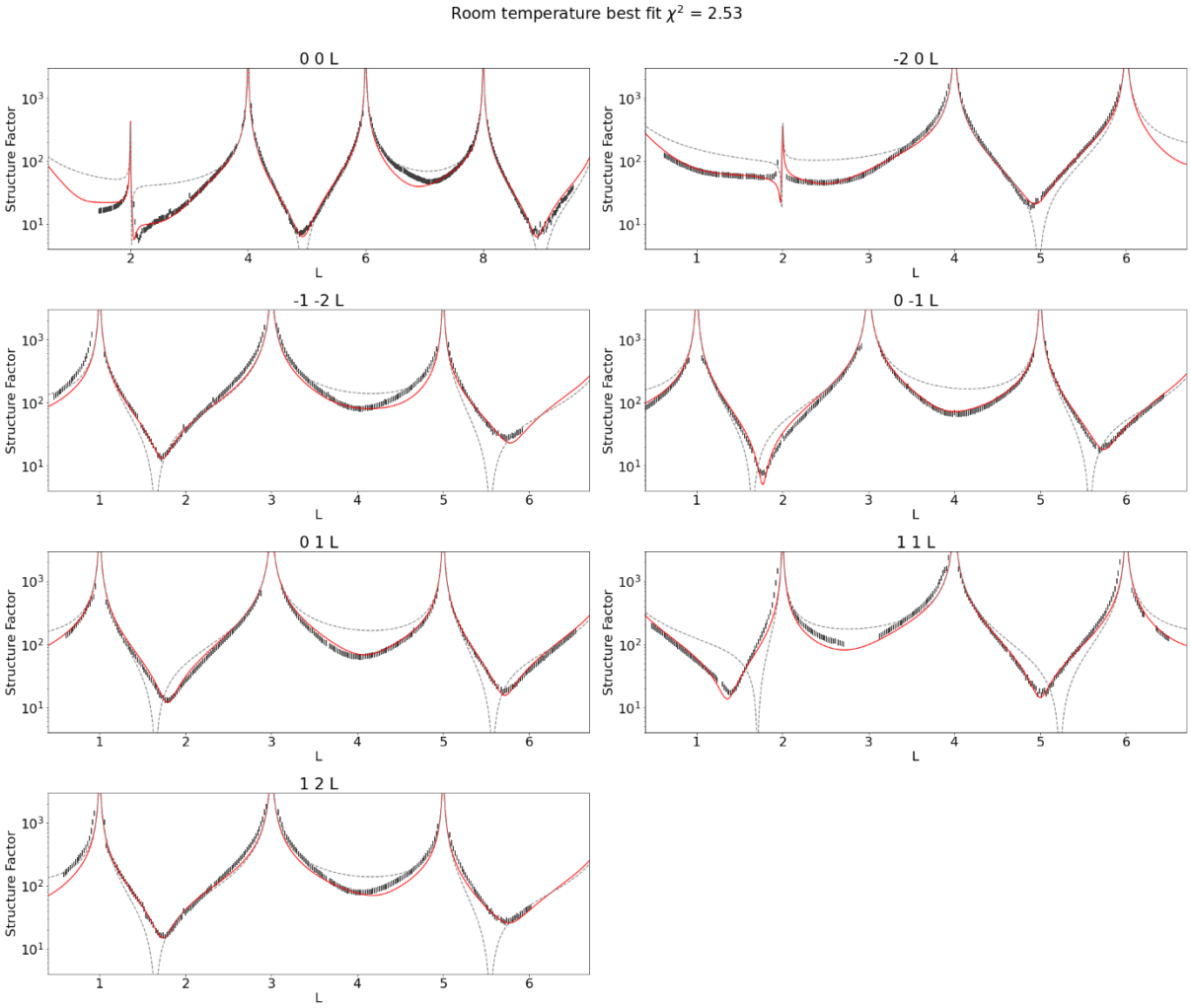


Table S2 – occupancies, starting bulk XYZ positions and final fit positions for top six atomic layers in High temperature best fit model. The X,Y,Z co-ordinates correspond to fractional positions along the a,b,c lattice directions.

El	Bulk X	Bulk Y	Bulk Z	Fit X	Error X	Fit Y	Error Y	Fit Z	Error Z	occval
O	0.0	0.0	1.0000	-0.0646	0.056	-0.0693	0.008	1.0500	0.006	0.16
La/Sr	0.5	0.5	0.9783	0.4523	0.001	0.4663	0.001	0.9855	0.001	0.16
Al	0.0	0.0	0.8370	-0.0238	0.010	-0.0001	0.002	0.8364	0.000	0.32
O	0.0	0.5	0.8370	0.0478	0.021	0.5397	0.006	0.8847	0.008	0.32
O	0.5	0.0	0.8370	0.4657	0.012	0.0400	0.006	0.8851	0.003	0.32
La/Sr	0.5	0.5	0.6957	0.4856	0.003	0.4878	0.001	0.7078	0.001	0.32
O	0.0	0.0	0.6740	0.0663	0.025	-0.0258	0.012	0.6669	0.001	0.32
O	0.5	0.5	0.5000	0.5122	0.002	0.5071	0.003	0.5040	0.001	0.94
La/Sr	0.0	0.0	0.4783	-0.0014	0.000	0.0086	0.000	0.4788	0.000	0.94
Al	0.5	0.5	0.3370	0.4829	0.004	0.5122	0.002	0.3330	0.000	0.97
O	0.5	0.0	0.3370	0.4994	0.007	-0.0204	0.003	0.3300	0.001	0.97
O	0.0	0.5	0.3370	-0.0327	0.002	0.4713	0.005	0.3289	0.003	0.97
La/Sr	0.0	0.0	0.1957	0.0021	0.000	0.0041	0.000	0.1952	0.000	1.00
O	0.5	0.5	0.1740	0.5043	0.002	0.5011	0.001	0.1758	0.002	1.0

Figure S2 – Comparison of CTR structure factor data (black error lines), best-fits (red lines) and bulk profiles (dotted grey lines) from full high temperature CTR dataset

