Supporting Information for

Transition from reconstruction towards thin film on the (110) surface of strontium titanate

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Supplemental Figure S1, showing LEED patterns of in (a) the 4×1 reconstruction and in (b) the $n \times 5$ reconstruction, with the 1×1 unit cell shown in red in both. Due to the much higher background in LEED compared to TED, as well as the much more dynamical nature of the scattering it is not clear in (b) whether it is a 1×5 or a 2×5 surface; as shown later in Supplemental Figure S5 and discussed in the main text the additional scattering from the "2" periodicity is weak due to disorder.



Supplemental Figure S2: (a-g) Map of the observed structures as a function of dosing of either Sr or Ti on the SrTiO₃ (110) surface, showing more involved, probably metastable, 2×4 and 6×4



structures. Note that the x-axis gives evaporation doses rather than coverage, which could be different due to diffusion of surface atoms into the bulk.

Supplemental Figure S3: STM image showing co-existing regions of (0,0,2) periodicity arrowed in an otherwise (0,0,1) periodicity.



Supplemental Figure S4: Ti 2*p* core-level photoemission spectra for the (4×1) (black curve) and (2×5) (red curve) reconstructed surfaces. The Ti valence is 4+ for both surfaces. The spectra are aligned and measured with photon energy of 605 eV at grazing angle.



Figure Supplemental S5: Pseudocolor representation of a filtered high-pass diffraction pattern of a 2×4 sample. In addition to the strong intensities of the 1×4 lattice, there is weak intensity correlating with а statistically disordered 2×4 superlattice, arrowed.

Н	Κ	Iobs	Icalc
3	3	58.69	61.37
2	6	99.63	124.41
3	6	31.16	91.2
1	7	287.76	248.97
2	7	91.72	101.18
4	10	35.44	33.79
3	11	63.07	59.64
4	11	59.44	46.8
1	13	72.11	71.65
2	13	109.61	72.86
4	13	24.11	89.24
0	14	360.47	372.52
1	14	53.92	62.78
2	14	177.06	171.9
4	14	43.46	43.93
0	15	841.4	890.74
1	15	80.94	58.6
2	15	431.31	488.2
3	15	24.23	51.45
4	15	106.42	89.87
0	17	64.16	65.68
4	18	162.92	104.76
0	19	62.49	62.7
1	19	61.85	65.21
0	21	40.61	74.72
2	22	84.35	56.02
2	23	128.22	118.68
3	23	1.49	44.91
1	26	0.51	51.67
2	26	0.28	58.18
1	29	38.48	48.51

Table S1: Experimental intensities and calculated intensities using Shelx for the 2×4 electron diffraction data using a kinematical model with occupational disorder.

Table S2: Experimental intensities and calculated intensities using Shelx for the 2×5 electron diffraction data using a kinematical model with occupational disorder.

Н	Κ	Iobs	Icalc
1	2	472.96	395.63

3	2	65.67	97.57
1	3	131.45	125.42
2	3	92.79	365.18
3	3	20.64	162.36
1	4	286.63	213.69
2	4	76.25	257.67
3	4	51.58	52.23
2	6	9.82	207.7
3	6	59.38	55.09
1	7	52.73	131.18
2	7	101.81	179.53
0	8	129.67	379.13
1	8	256.9	339.86
2	8	38.48	211.07
3	8	81.32	174.62
0	9	794.39	552.22
1	9	1483.67	1276.86
2	9	222.53	292.65
3	9	257.23	455.64
1	12	233.43	103.16
2	12	370.19	161.19
3	12	62.2	116.23
0	13	377.21	273.44
1	13	265.06	164.55
2	13	174.82	243.11
1	14	113.94	125.41
2	14	14.7	194.71
3	14	37.78	80.24
0	16	345.04	334.51
1	16	93.59	84.72
2	16	180.97	161.17
3	16	35.16	49.7
0	17	704.56	850.27
1	17	155.24	147.37
2	17	387.72	350.93
3	17	58.54	745.76
0	18	1293.38	1535.41
1	18	166.37	121.64
2	18	736.46	700.02
3	18	68.84	81.34
0	19	3710.21	5040.02

1	19	213.31	246.48
2	19	2184.5	2379.52
1	21	79.84	73.14
2	21	151.99	78.1
0	22	222.85	138.72
1	22	77.69	209.62
0	23	97.81	253.09
1	23	90.54	89.75
2	23	66.38	137.92
1	24	80.12	77.79
2	24	129.79	100.23
0	26	349.96	192.53
1	26	69.78	108.3
0	27	70.15	52.81