Table SI 1: Structural parameters for $La_{2-x}Sr_xCoTiO_6$ ($0 \le x \le 0.5$) oxides obtained from SXRD/XRD and NPD data; agreement factors are given for NPD data.

	[b] La_2CoTiO_6	[c] $La_{1.90}Sr_{0.10}CoTiO_6$	[d] $La_{1.80}Sr_{0.20}CoTiO_6$	[e] $La_{1.70}Sr_{0.30}CoTiO_6$	[f] $La_{1.50}Sr_{0.50}CoTiO_6$
X-ray Source	Synchrotron	Lab apparatus	Synchrotron	Synchrotron	Synchrotron
Space Group[a]	$P2_1/n$	$P2_1/n$	$P2_1/n$	Pnma	Pnma
***a* (Å)**	5.55510 (9)	5.55318(8)	5.5502(11)	5.52939(10)	5.49186(7)
***b* (Å)**	5.57813 (8)	5.56557(7)	5.54838(9)	7.81873(13)	7.77013(10)
c (Å)	7.85718 (12)	7.84995(10)	7.83763(10)	5.54331(9)	5.53014(7)
β (deg)	90.005 (5)	90.007(9)	90.014(7)		
Volume (Å^3)	243.47 (1)	242.62 (1)	241.36 (1)	239.65 (1)	235.99 (1)
La/Sr position	4e	4e	4e	4c	4c
Occ La/Sr	1/0	0.92(1)/0.08 (1)	0.89(1)/0.11(1)	0.423(2)/0.077(2)	0.373(4)/0.127(4)
x	0.0059(3)	0.0056(4)	0.0048(4)	0.02348(16)	0.0092(3)
y	−0.03335 (16)	-0.03105(18)	-0.0271(2)	¼	¼
z	0.2494 (7)	0.2494(10)	0.2503(16)	0.0037(4)	0.0021(2)
U*100 (Å^2)	0.55(2)	0.66(2)	0.75 (2)	0.622(17)	0.989(17)
B' position	2d	2d	2d	4b	4b
Occ Co/Ti	0.453 (3)/0.047(3)	0.413(4)/0.087(4)	0.287(9)/0.213(9)	0.250/0.250	0.250/0.250
U*100 (Å^2)	0.05(8)	0.36(3)	0.7(8)	0.14(5)	0.60(8)
B'' position	2c	2c	2c		
Occ Co/Ti	0.047(3)/0.453(3)	0.087(4)/0.413(4)	0.213(9)/0.287(9)		
U*100 (Å^2)	0.38 (8)	0.7(2)	0.7(3)		
O(1) position	4e	4e	4e	8d	8d
x	-0.7229(9)	-0.7234(8)	-0.7219(11)	0.7774(3)	0.7631(3)
y	0.7041(9)	0.7081(8)	0.7228(11)	0.0348 (2)	0.03058 (11)
z	0.0381(9)	0.0375(10)	0.0387(10)	0.2228 (3)	0.2374(3)
Occ	1.00	1.00	1.00	1.000	1.000
U*100 (Å^2)	0.58(11)	0.62(11)	1.4(2)	0.76(3)	1.25(2)
O(2) position	4e	4e	4e	4c	4c
x	-0.2058(9)	-0.2102(8)	-0.2173(11)	0.0084(4)	0.0029(5)
y	0.7759(9)	0.7757(8)	0.7849(11)	3/4	3/4
z	-0.0404(9)	-0.0390(10)	-0.0351(10)	0.4342(4)	0.4426(2)
Occ	1.00	1.00	1.00	0.500	0.500
U*100 (Å^2)	0.51(11)	0.82 (12)	0.49(16)	0.78(4)	0.91(3)
O(3) position	4e	4e	4e		
x	-0.0740(5)	-0.0725(4)	-0.06989(4)		
y	0.5143(4)	0.5128(3)	0.51043(5)		
z	0.2473(8)	0.2475(3)	0.2510(15)		
Occ	1.00	1.00	1.00		
U*100 (Å^2)	0.83 (5)	0.76 (4)	0.72(4)		

[a] $P2_1/n$: 4e (xyz), 2d (½ 0 0), 2c (0 ½ 0); Pnma: 4c (x ¼ z), 4b (0 0 ½), 8d (xyz)

[b] χ^2= 2.34, R_{wp}= 5.89%, R_{exp}= 3.85%, R_B= 3.24%,

[c] χ^2= 3.10, R_{wp}= 4.69%, R_{exp}=2.66%, R_B= 2.30%,

[d] χ^2= 2.97, R_{wp}= 4.53%, R_{exp}= 2.63%, R_B= 2.52%,

[e] χ^2= 2.64, R_{wp}= 4.44%, R_{exp}= 2.74%, R_B= 2.08%,

[f] χ^2= 6.43, R_{wp}= 4.04%, R_{exp}= 1.59%, R_B= 2.70%,

Table SI 2: Selected structural information for $La_{2-x}Sr_xCoTiO_6$ ($0 \leq x \leq 0.5$) oxides obtained from XRD and NPD data. Angles are given in degrees and distances in Å.

	La_2CoTiO_6	$La_{1.90}Sr_{0.10}CoTiO_6$	$La_{1.80}Sr_{0.20}CoTiO_6$	[a] $La_{1.70}Sr_{0.30}CoTiO_6$	[a] $La_{1.50}Sr_{0.50}CoTiO_6$
[b] **Tilt angle φ**	16.9(1)	16.6(1)	15.4(1)	15.0(1)	13.1(1)
[c] **Tilt angle θ**	16.9(1)	16-6(1)	15.4(1)	15.0(1)	13.1(1)
[d] **Tilt angle μ**	11.7(1)	118(1)	11.2(1)	10.0(1)	7.5(1)
d B'-O(1) x 2	2.085(5)	2.068(5)	1.994(6)	1.9880(17)	1.9667(16)
d B'-O (2) x 2	2.082(5)	2.062(5)	1.991(6)	1.9875(17)	1.9642(16)
d B'-O (3) x 2	2.029(6)	2.026(2)	1.989(12)	1.9891(4)	1.9684(2)
Average B'-O (Co-O)	2.065(2)	2.052 (1)	1.991 (4)	1.9882 (6)	1.9664(5)
B''-O(1) x 2	1.938(5)	1.949(5)	2.001(6)		
B''-O(2) x 2	1.943(5)	1.955 (5)	2.007(6)		
B''-O(3) x 2	1.988(6)	1.988 (2)	2.005 (12)		
Average B''-O (Ti-O)	1.956(3)	1.964(1)	2.004 (4)		
d A-O(1)	2.678(7) 2.448(7) 3.309(7) 2.769(8)	2.676(8) 2.459(8) 3.284(8) 2.779(9)	2.642 (11) 2.471 (11) 3.232(11) 2.806 (12)	2.482(2) x 2 2.665(2) x 2 2.7833(19) x 2 3.186(2) x 2	2.535117) x 2 2.6318(17) x 2 2.8409(15) x 2 3.0219(16) x 2
d A-O(2)	2.775(8) 3.314(7) 2.448(7) 2.669(7)	2.782(9) 3.287(1) 2.461(8) 2.669(8)	2.759 (13) 3.245 (11) 2.458 (11) 2.693 (10)	2.433(3) 3.121(3) 2.966(2) 2.617(2)	2.4602(16) 3.0716(16) 2.832(3) 2.700(3)
A-O(3)	3.087(2) 2.562(2) 3.167(3) 2.414(3)	3.060(2) 2.5795(4) 3.160(3) 2.421(3)	3.0103(12) 2.5984(12) 3.138(3) 2.428(3)		
Coordination A-site d (A-O) ≤ 3 Å	8	8	8	9	9
[a] **d A-B'**	3.377(5) 3.433(4) 3.264(4)	3.375(6) 3.426(4) 3.270(5)	3.378(5) 3.420(2) 3.276(3)	3.4932(7) 3.2804(7) 3.411(2) 3.377(2)	3.4050(14) 3.3225(13) 3.3891(9) 3.3701 9)
[a] **d A-B''**	3.259(4) 3.381(4) 3.438(5)	3.261(5) 3.383(5) 3.435(5)	3.273(3) 3.380(2) 3.424(5)		

[a] For S.G. Pnma only a B-site exists with co-ordination: B'-O(1) x 2, B'-O(2) x 2 and B'-O(2)' x 2
[b] With [110] for $P2_1/n$; with [101] for Pnma
[c] With [1-10] for $P2_1/n$; with [10-1] for Pnma
[d] With [001] for $P2_1/n$; with [010] for Pnma

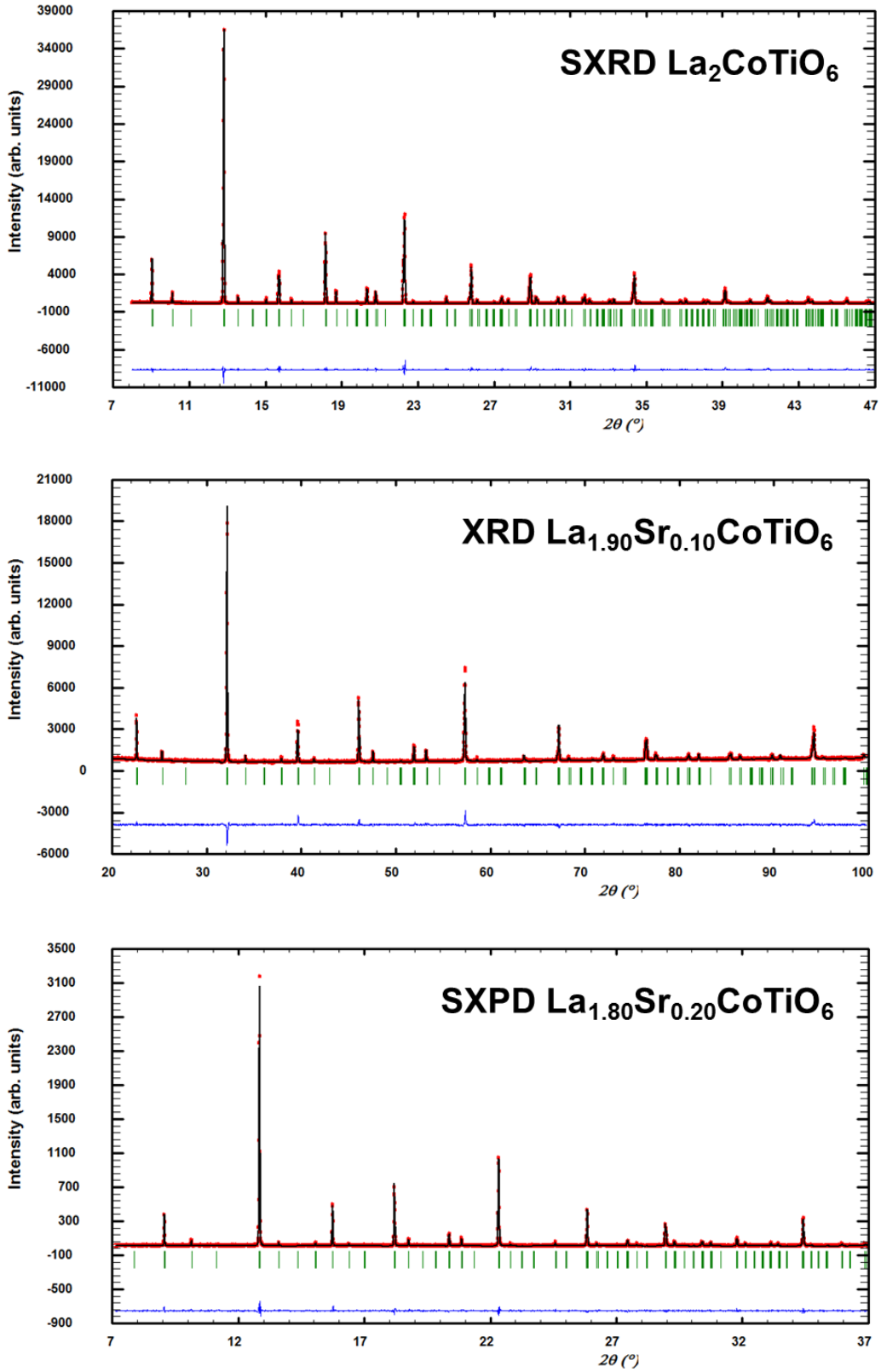
Table SI 3: Rietveld refinement results with AMPLIMODES for S.G $P2_1/n$

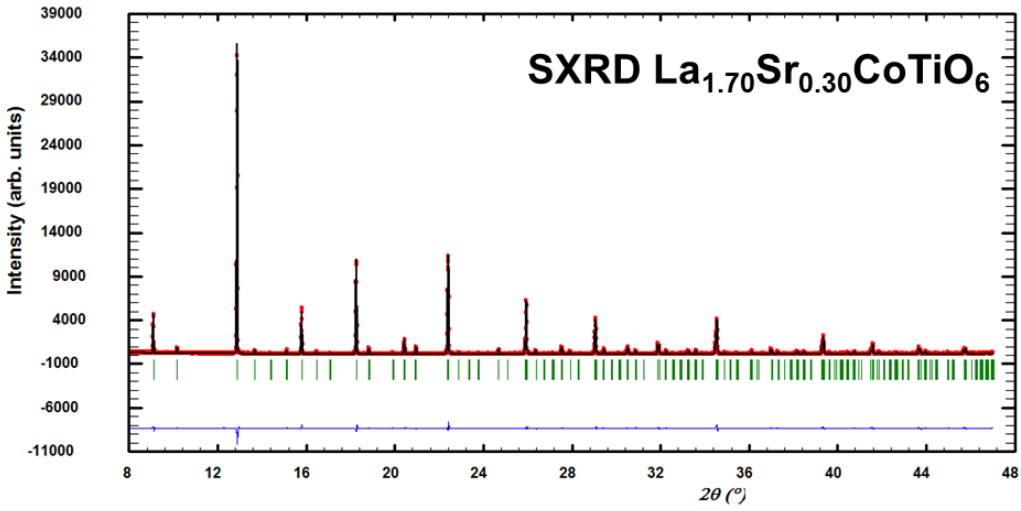
Mode/Atom	La_2CoTiO_6 (Å)	$La_{1.90}Sr_{0.10}CoTiO_6$ (Å)	$La_{1.80}Sr_{0.20}CoTiO_6$ (Å)
A1-GM1+ (O)	0.003(8)	0.040 (7)	0.020 (2)
A2-GM3+ (O)	-0.08 (1)	-0.062 (1)	-0.02 (2)
A3-GM4+ (O)	-1.199 (4)	-1.171 (3)	-1.110 (3)
A4-GM5+ (La/Sr)	0.066 (4)	0.063(5)	0.052 (5)
A5-GM5+ (La/Sr)	0.01 (1)	0.01 (2)	0.00 (2)
A6-GM5+ (O)	0.02 (1)	0.02 (1)	-0.04(3)
A7-GM5+ (O)	-0.036 (7)	-0.029 (5)	-0.042 (7)
A8-X2+ (O)	0.003 (6)	0.007 (6)	0.019 (8)
A9-X3+ (O)	-0.796 (5)	-0.744 (4)	-0.691 (4)
A10-X5+ (La/Sr)	-0.371 (2)	-0.346(2)	-0.296 (3)
A11-X5+ (O)	-0.03(2)	-0.02 (2)	0.03(3)
A12-X5+ (O)	0.159 (4)	0.141 (4)	0.1171 (7)

Table SI 4: Rietveld refinement results with AMPLIMODES for S.G Pnma

Mode/Atom	$La_{1.70}Sr_{0.30}CoTiO_6$ (Å)	$La_{1.50}Sr_{0.50}CoTiO_6$ (Å)
A1-R4+ (O)	-1.085 (3)	-0.923 (2)
A2-R5+ (La/Sr)	0.042 (4)	0.023 (2)
A3-R5+ (O)	-0.030 (6)	-0.030 (3)
A4-X5+(La/Sr)	0.266 (2)	0.102 (3)
A5-X5+ (O)	0.095 (5)	0.031 (5)
A6-M2+ (O)	0.002 (5)	0.005 (5)
A7-M3+ (O)	0.6184 (6)	0.284 (4)

Figure SI 1: Experimental (points), calculated (continuous line) SR-HRPD and XRD patterns and their difference (bottom) for $La_{2-x}Sr_xCoTiO_6$ ($0 \leq x \leq 0.5$) oxides. The row of vertical bars indicates the positions of Bragg peaks.




SXRD La1.70Sr0.30CoTiO6
Intensity (arb. units)
39000
34000
29000
24000
19000
14000
9000
4000
-1000
-6000
-11000
8
12
16
20
24
28
32
36
40
44
48
2θ (°)

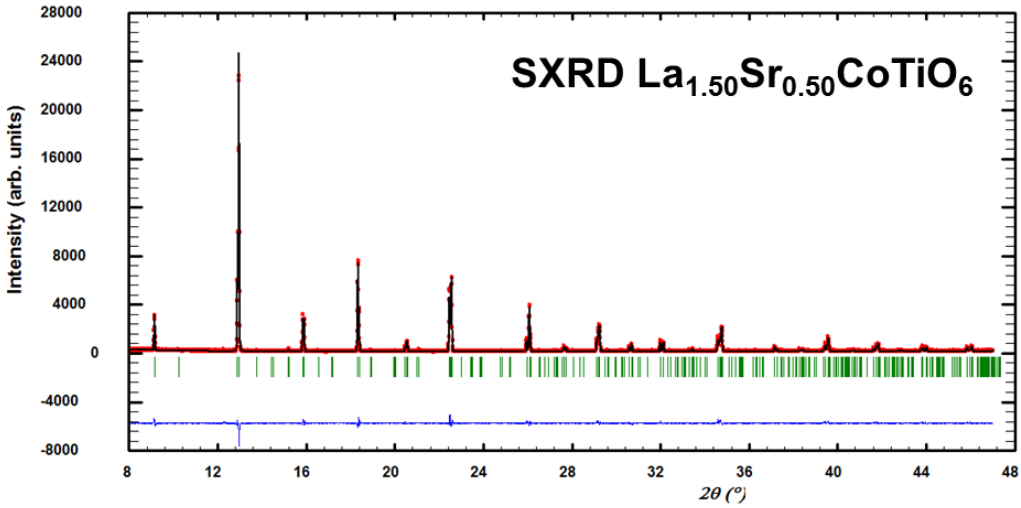
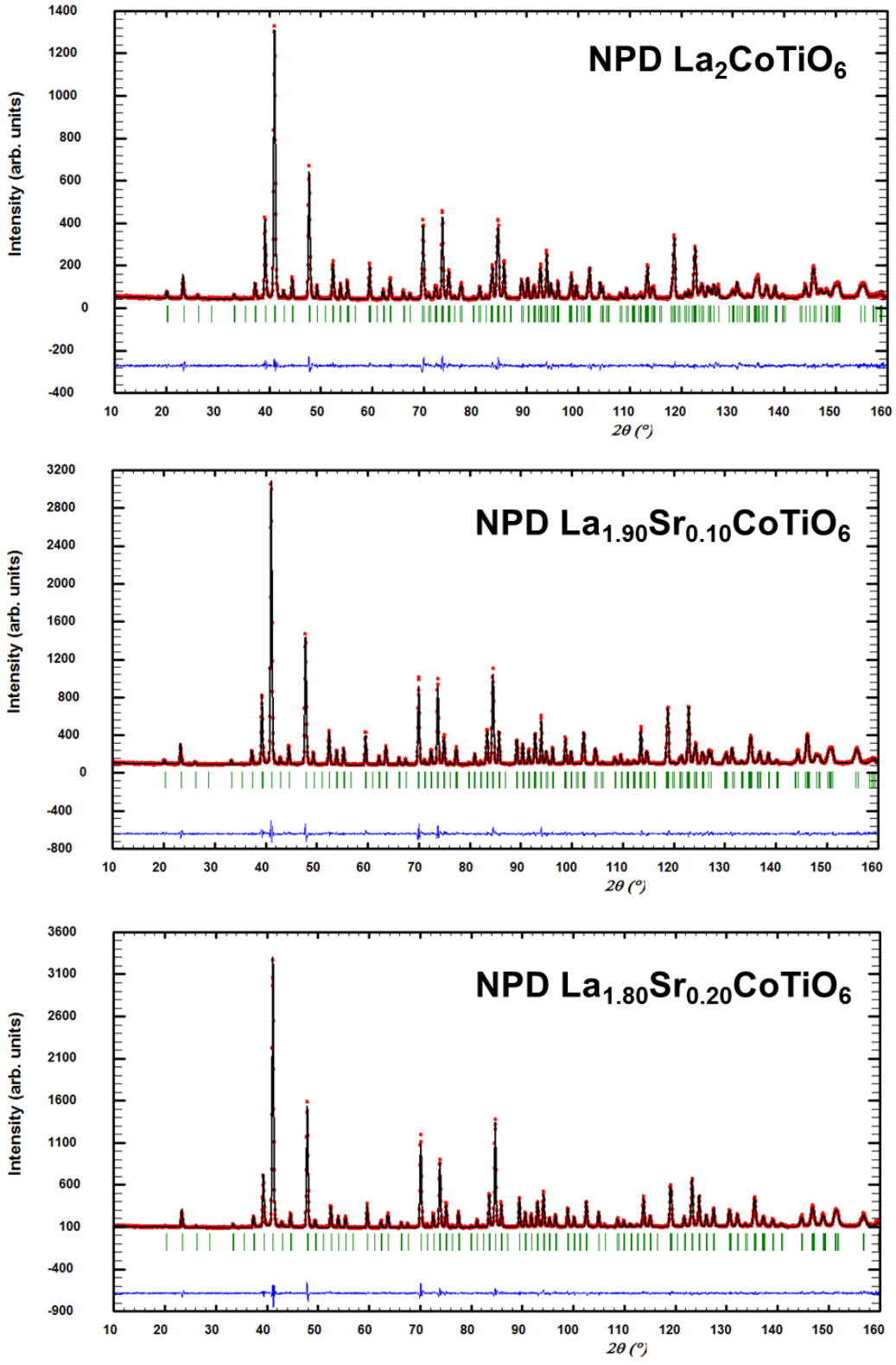
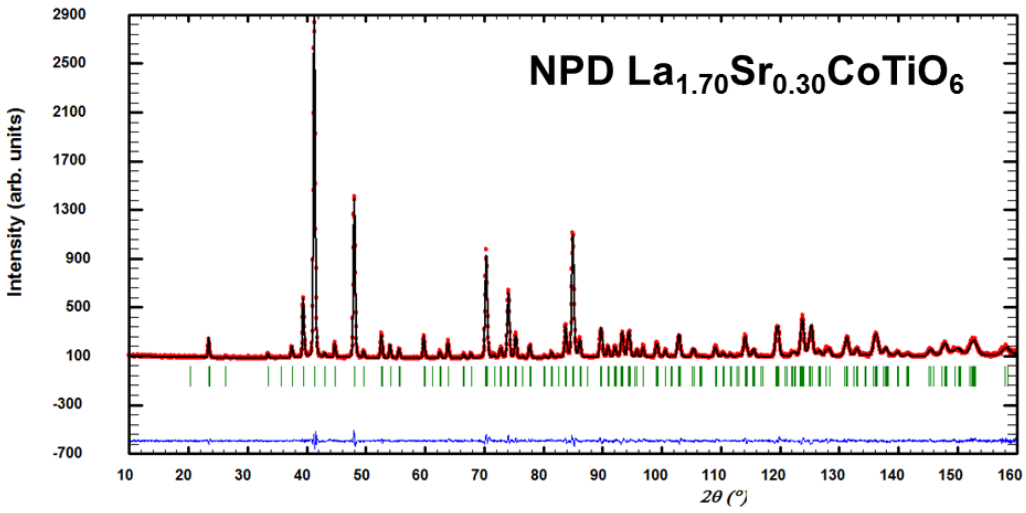
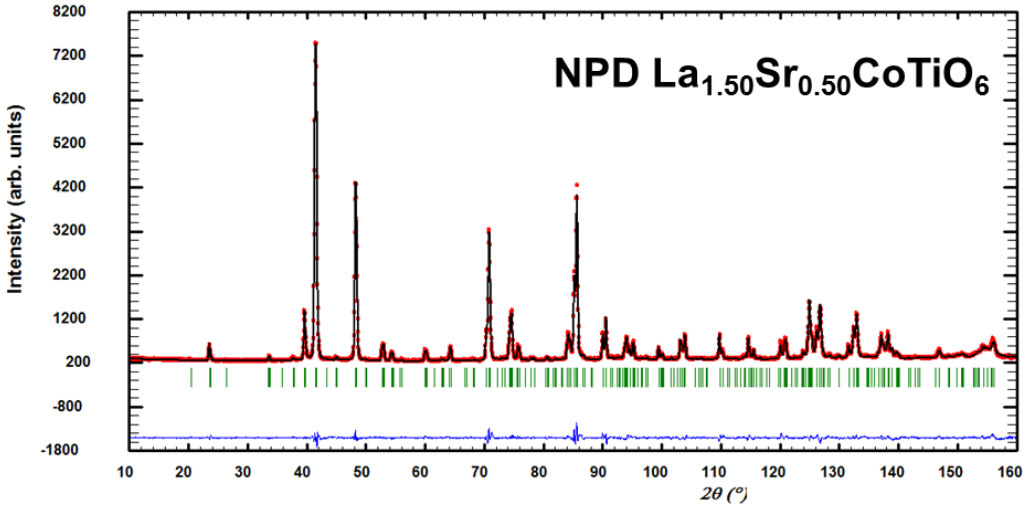

SXRD La1.50Sr0.50CoTiO6
Intensity (arb. units)
28000
24000
20000
16000
12000
8000
4000
0
-4000
-8000
8
12
16
20
24
28
32
36
40
44
48
2θ (°)

Figure SI 2: Experimental (points), calculated (continuous line) NPD patterns and their difference (bottom) for $La_{2-x}Sr_xCoTiO_6$ ($0 \leq x \leq 0.5$) oxides. The row of vertical bars indicates the positions of Bragg peaks.




NPD La1.70Sr0.30CoTiO6
Intensity (arb. units)
2900
2500
2100
1700
1300
900
500
100
-300
-700
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
2θ (°)


NPD La1.50Sr0.50CoTiO6
Intensity (arb. units)
8200
7200
6200
5200
4200
3200
2200
1200
200
-800
-1800
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
2θ (°)