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Supplementary Materials

Oxygen transport pathways in Ruddlesden-Popper structured oxides revealed via in-situ neutron diffraction

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Figure S1. Fourier observed nuclear density diagram at 1070 K with a pO_2 of .1 atm for $LaSrCo_{0.5}Fe_{0.5}O_{4-\delta}$ (RPn1) at an isosurface level of 0.15, 1.3% of maximum. The progression of color from blue to green to red denotes an increase in the observed nuclear density, showing the probable positions for the atoms.

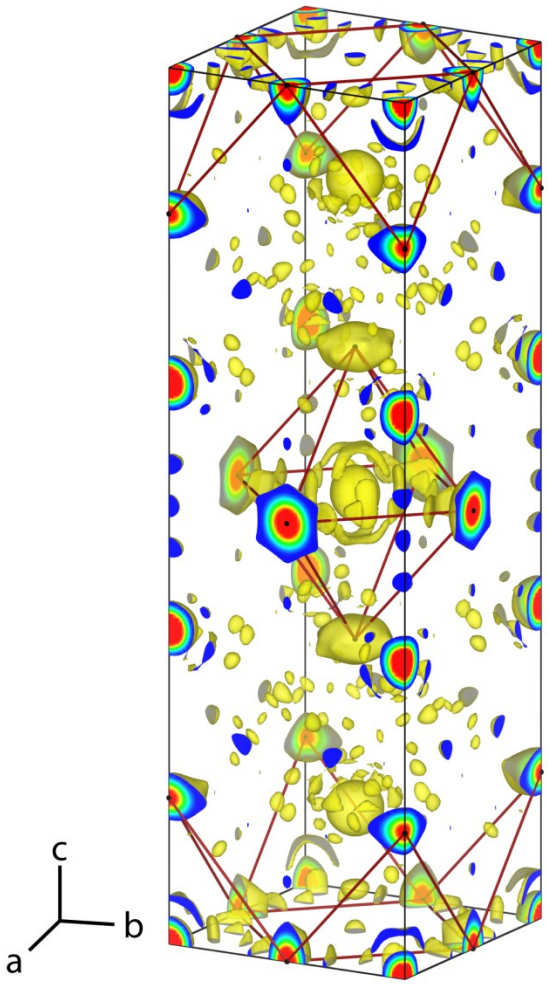

Figure S2. Fourier observed nuclear density diagram at 1070 K with a pO_2 of .1 atm for $La_{0.3}Sr_{2.7}CoFeO_{7-\delta}$ (RPn2) at an isosurface level of 0.2, 2.1% of maximum. The progression of color from blue to green to red denotes an increase in the observed nuclear density, showing the probable positions for the atoms.

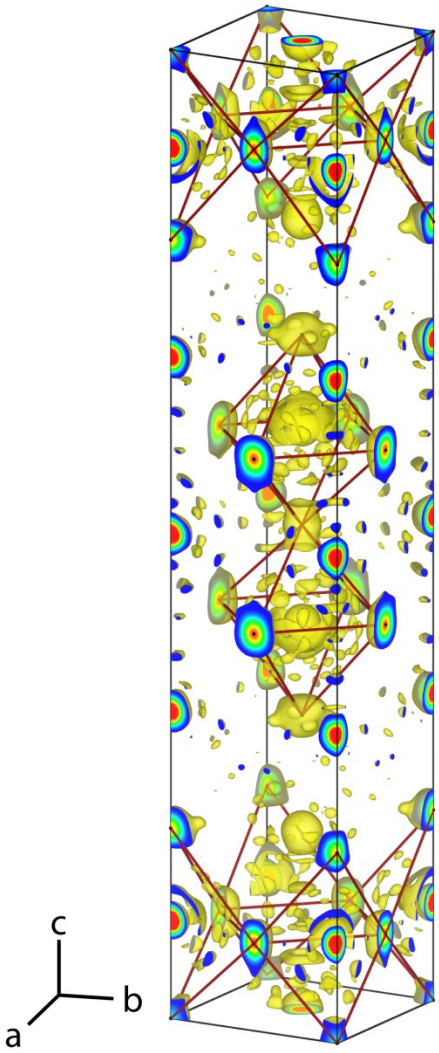

Figure S3. Fourier observed nuclear density diagram at 1070 K with a pO_2 of .1 atm $LaSr_3Co_{1.5}Fe_{1.5}O_{10-\delta}$ (RPn3) at an isosurface level of 0.25, 2.4% of maximum. The progression of color from blue to green to red denotes an increase in the observed nuclear density, showing the probable positions for the atoms.

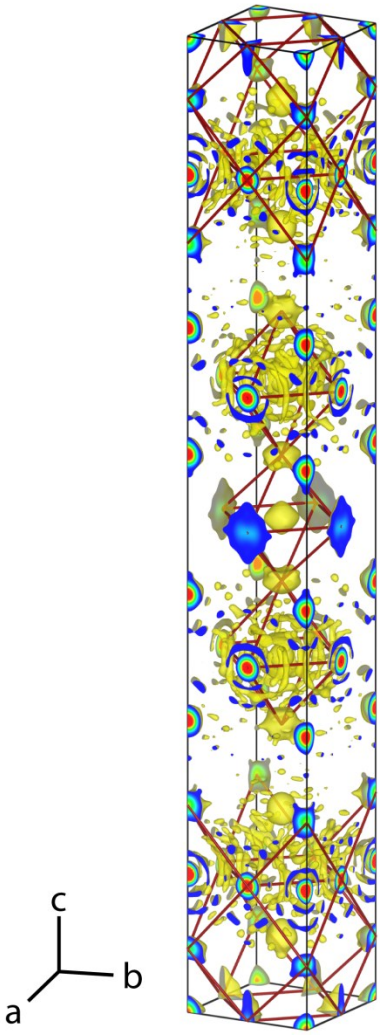

Figure S4. Fourier observed nuclear density diagram at 1070 K with a pO_2 of .1 atm for $LaSrCo_{0.5}Fe_{0.5}O_{4-\delta}$ (RPn1) at an isosurface level of 0.10, 0.9% of maximum. The progression of color from blue to green to red denotes an increase in the observed nuclear density, showing the probable positions for the atoms.

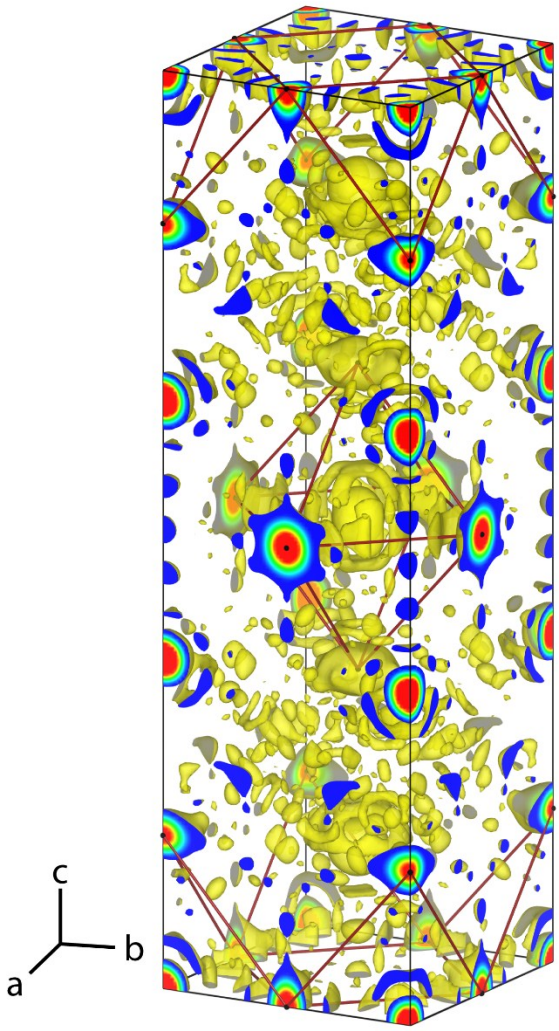

Figure S5. Fourier observed nuclear density diagram at 1070 K with a pO_2 of .1 atm for $La_{0.3}Sr_{2.7}CoFeO_{7-\delta}$ (RPn2) at an isosurface level of 0.12, 1.3% of maximum. The progression of color from blue to green to red denotes an increase in the observed nuclear density, showing the probable positions for the atoms.

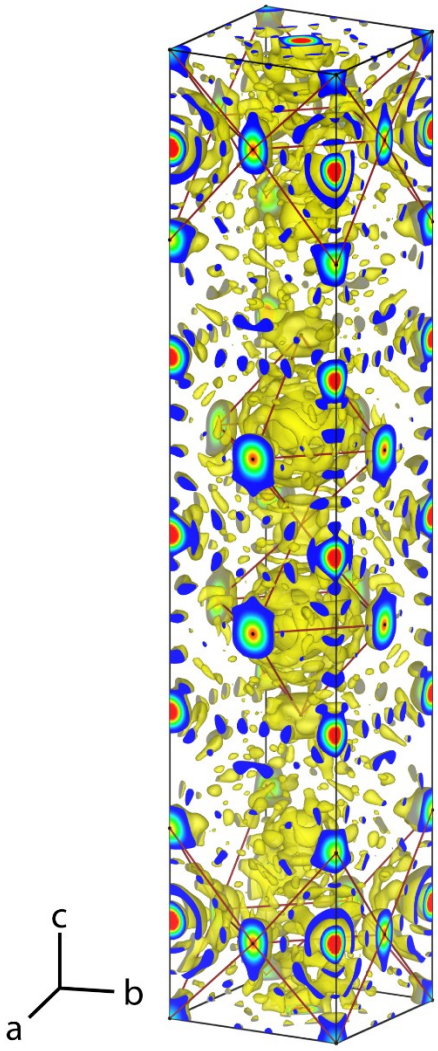

Figure S6. Fourier observed nuclear density diagram at 1070 K with a pO_2 of .1 atm $LaSr_3Co_{1.5}Fe_{1.5}O_{10-\delta}$ (RPn3) at an isosurface level of 0.25, 1.4% of maximum. The progression of color from blue to green to red denotes an increase in the observed nuclear density, showing the probable positions for the atoms.

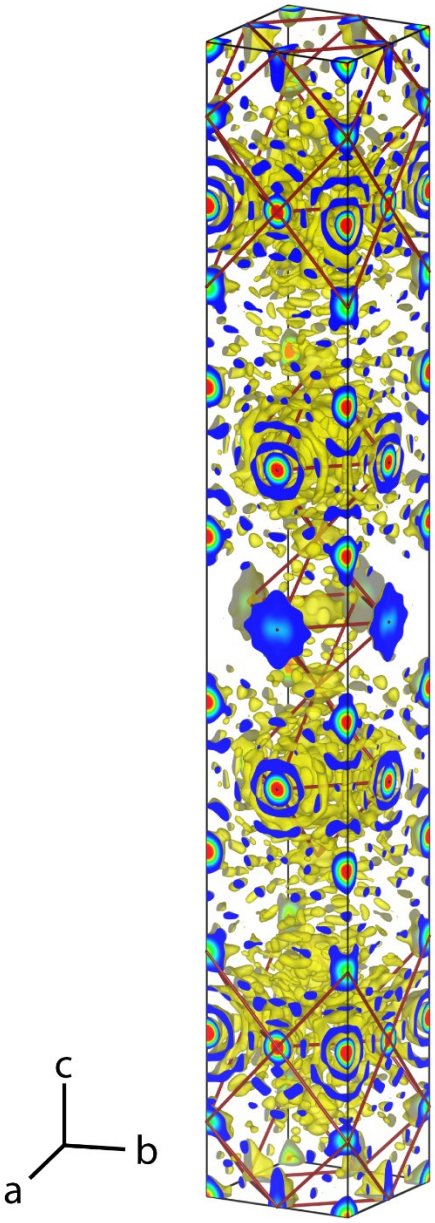

Table S1: Tetragonal space group I4/mmm, Rietveld fit parameters for RPn1 structure at 765K, 865K, and 967 K with sites:

RPn1: La/Sr1: 4e (0,0,Z), Co/Fe1: 2a (0,0,0), O1: 4e (0,0,Z), O2: 4c (0.5,0,0)

$LaSrCo_{0.5}Fe_{0.5}O_{3.942}$ (RPn1) I4/mmm 765 K

a,b (Å)	3.859 (1)	χ^2	4.922
c (Å)	12.738(1)	R_P %	0.1182
V (Å^3)	189.66(1)	wR_P %	0.0461
ρ_c (g/cm^3)	6.092		

	Atomic displacement parameters (Å^2)*100				Fractional
	U_{11}/U_{iso}	U_{22}	U_{33}	Z	Occupancy
La/Sr1	1.40(2)			0.359(1)	
Co/Fe1	1.17(3)				
O1	3.09(4)	3.09(4)	1.46(6)	0.166(1)	1.0
O2	0.64(5)	1.99(6)	2.79(8)		1.0

$LaSrCo_{0.5}Fe_{0.5}O_{3.942}$ (RPn1) I4/mmm 865 K

a,b (Å)	3.863 (1)	χ^2	4.674
c (Å)	12.789(1)	R_P %	0.1139
V (Å^3)	190.84(1)	wR_P %	0.0441
ρ_c (g/cm^3)	6.055		

	Atomic displacement parameters (Å^2)*100				Fractional
	U_{11}/U_{iso}	U_{22}	U_{33}	Z	Occupancy
La/Sr1	1.60(2)			0.359(1)	
Co/Fe1	1.34(3)				
O1	3.49(5)	3.49(5)	1.63(7)	0.167(1)	1.0
O2	0.86(5)	2.32(7)	2.87(9)		1.0

$LaSrCo_{0.5}Fe_{0.5}O_{3.942}$ (RPn1) I4/mmm 967 K

a,b (Å)	3.868 (1)	χ^2	4.522
c (Å)	12.840(1)	R_P %	0.1055
V (Å^3)	192.08(1)	wR_P %	0.0423
ρ_c (g/cm^3)	5.999		

	Atomic displacement parameters (Å^2)*100				Fractional
	U_{11}/U_{iso}	U_{22}	U_{33}	Z	Occupancy
La/Sr1	1.90(2)			0.359(1)	
Co/Fe1	1.53(3)				
O1	3.52(7)	3.52(7)	1.8(1)	0.167(1)	0.971(8)
O2	1.0(1)	2.7(1)	3.3(1)		1.0

Table S2: Tetragonal space group I4/mmm, Rietveld fit parameters for RPn2 structure at 765K, 865K, and 967 K with sites:

RPn2: La/Sr1: 4e (0,0,Z), La/Sr2: 2b (0,0,0.5), Co/Fe1: 4e (0,0,Z), O1: 4e (0,0,Z), O2: 8g (0,0.5,Z), O3: 2a (0,0,0)

$La_{0.3}Sr_{2.7}CoFeO_{6.378}$ (RPn2) I4/mmm 765 K

a,b (Å)	3.888(1)	χ^2	5.333
c (Å)	20.306(1)	R_P %	0.1677
V (Å^3)	306.97(1)	wR_P %	0.0678
ρ_c (g/cm^3)	5.382		

	Atomic displacement parameters (Å^2)*100				Fractional
	U_{11}/U_{iso}	U_{22}	U_{33}	Z	Occupancy
La/Sr1	2.22(4)			0.317(1)	
La/Sr2	1.93(6)				
Co/Fe1	1.33(4)			0.100(1)	
O1	2.92(6)			0.195(1)	1.0
O2	2.43(9)	1.40(8)	3.6(1)	0.092(1)	0.956(6)
O3	3.1(2)				0.70(1)

$La_{0.3}Sr_{2.7}CoFeO_{6.378}$ (RPn2) I4/mmm 865 K

a,b (Å)	3.900(1)	χ^2	4.918
c (Å)	20.356(1)	R_P %	0.1532
V (Å^3)	309.59(1)	wR_P %	0.0637
ρ_c (g/cm^3)	5.315		

	Atomic displacement parameters (Å^2)*100				Fractional
	U_{11}/U_{iso}	U_{22}	U_{33}	Z	Occupancy
La/Sr1	2.60(5)			0.317(1)	
La/Sr2	2.25(7)				
Co/Fe1	1.61(4)			0.100(1)	
O1	3.32(6)			0.195(2)	1.0
O2	2.9(1)	1.53(9)	3.9(1)	0.091(1)	0.941(6)
O3	3.8(3)				0.64(2)

$La_{0.3}Sr_{2.7}CoFeO_{6.378}$ (RPn2) I4/mmm 967 K

a,b (Å)	3.912(1)	χ^2	4.529
c (Å)	20.409(1)	R_P %	0.1459
V (Å^3)	312.28(1)	wR_P %	0.0587
ρ_c (g/cm^3)	5.265		

	Atomic displacement parameters (Å^2)*100				Fractional
	U_{11}/U_{iso}	U_{22}	U_{33}	Z	Occupancy
La/Sr1	2.87(6)			0.317(1)	
La/Sr2	2.52(7)				
Co/Fe1	1.76(5)			0.101(1)	
O1	3.65(7)			0.196(2)	1.0
O2	3.4(1)	1.8(1)	4.4(2)	0.091(1)	0.942(6)
O3	4.7(3)				0.61(2)

Table S3: Tetragonal space group I4/mmm, Rietveld fit parameters RPn3 structure at 765K, 865K, and 967 K with sites:

RPn3: La/Sr1: 4e(0,0,Z), La/Sr2: 4e (0,0,Z), Co/Fe1: 4e (0,0,Z), Co/Fe2: 2a (0,0,0), O1: 4e (0,0,Z), O2: 8g (0,0.5,Z), O3: 4e (0,0,Z), O4: 4c (0,0.5,0)

$LaSr_3Co_{1.5}Fe_{1.5}O_{9.582}$ (RPn3) I4/mmm 765 K

a,b (Å)	3.879(1)	χ^2	3.064
c (Å)	28.235(1)	R_P %	0.1407
V (Å^3)	424.90(1)	wR_P %	0.0546
ρ_c (g/cm^3)	5.737		

	Atomic displacement parameters (Å^2)*100				Fractional
	U_{11}/U_{iso}	U_{22}	U_{33}	Z	Occupancy
La/Sr1	1.9(1)			0.2013(1)	
La/Sr2	2.0(1)			0.0696(2)	
Co/Fe1	1.1(1)			0.1398(1)	
Co/Fe2	1.5(1)				
O1	2.7(2)			0.2106(2)	1.0
O2	2.0(2)			0.1378(1)	1.0
O3	3.5(3)			0.0675(3)	1.0
O4	5.3(4)	1.4(2)	7.3(5)		1.0

$LaSr_3Co_{1.5}Fe_{1.5}O_{9.582}$ (RPn3) I4/mmm 865 K

a,b (Å)	3.889(1)	χ^2	2.834
c (Å)	28.366(1)	R_P %	0.1260
V (Å^3)	429.09(2)	wR_P %	0.0505
ρ_c (g/cm^3)	5.65.3		

	Atomic displacement parameters (Å^2)*100				Fractional
	U_{11}/U_{iso}	U_{22}	U_{33}	Z	Occupancy
La/Sr1	2.1(1)			0.2013(1)	
La/Sr2	2.4(1)			0.0704(2)	
Co/Fe1	1.4(1)			0.1404(1)	
Co/Fe2	1.9(1)				
O1	3.4(1)			0.2113(2)	1.0
O2	2.2(1)			0.1380(2)	0.98(1)
O3	4.1(3)			0.0664(3)	0.99(2)
O4	7.9(7)	1.6(3)	8.8(7)		0.94(2)

$LaSr_3Co_{1.5}Fe_{1.5}O_{9.582}$ (RPn3) I4/mmm 967 K

a,b (Å)	3.900(1)	χ^2	2.786
c (Å)	28.523(1)	R_P %	0.1577
V (Å^3)	433.78(2)	wR_P %	0.0587
ρ_c (g/cm^3)	5.568		

	Atomic displacement parameters (Å^2)*100				Fractional
	U_{11}/U_{iso}	U_{22}	U_{33}	Z	Occupancy
La/Sr1	2.5(1)			0.2013(1)	
La/Sr2	2.8(1)			0.0710(2)	
Co/Fe1	1.5(1)			0.1409(1)	
Co/Fe2	2.7(1)				
O1	3.8(2)			0.2114(2)	1.0
O2	2.5(1)			0.1380(2)	0.98(1)
O3	4.7(3)			0.0654(3)	0.95(2)
O4	12(1)	2.2(4)	11.4(9)		0.89(4)