Electronic Supplementary Material (ESI) for Dalton Transactions.

SUPPLEMENTARY INFORMATION FOR

Complex magnetic behaviour of $Sr_2CoNb_{1-x}Ti_xO_6$ ($0 \leq x \leq 0.5$) as a result of a flexible microstructure

M. Teresa Azcondo[a,], Julio Romero de Paz[b], Khalid Boulahya[c], Clemens Ritter[d], Flaviano García-Alvarado[a] and Ulises Amador[a]*

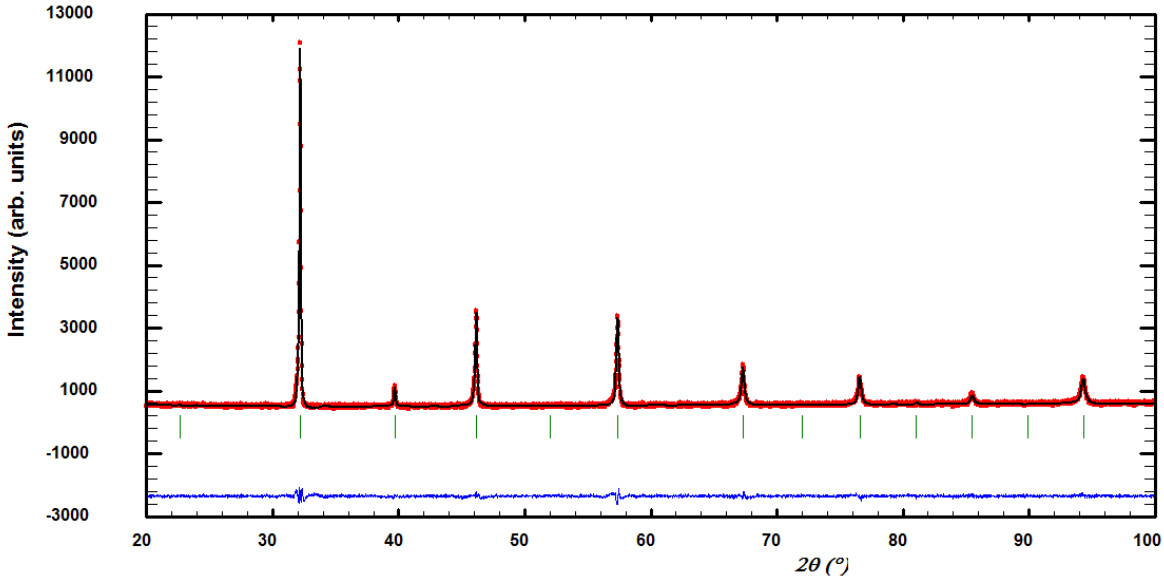

Figure SI 1. Experimental (red circles) and calculated (black continuous line) XRD patterns (and their difference, blue line at the bottom) for Sr_2CoNbO_6.

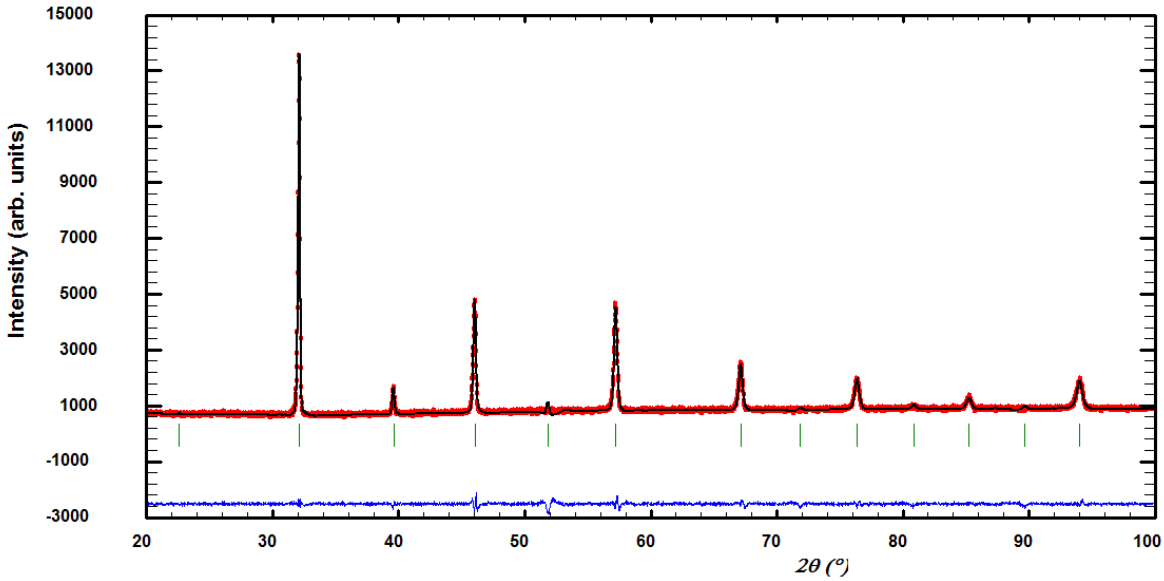

Figure SI 2. Experimental (red circles) and calculated (black continuous line) XRD patterns (and their difference, blue line at the bottom) for $Sr_2CoNb_{0.90}Ti_{0.10}O_6$.

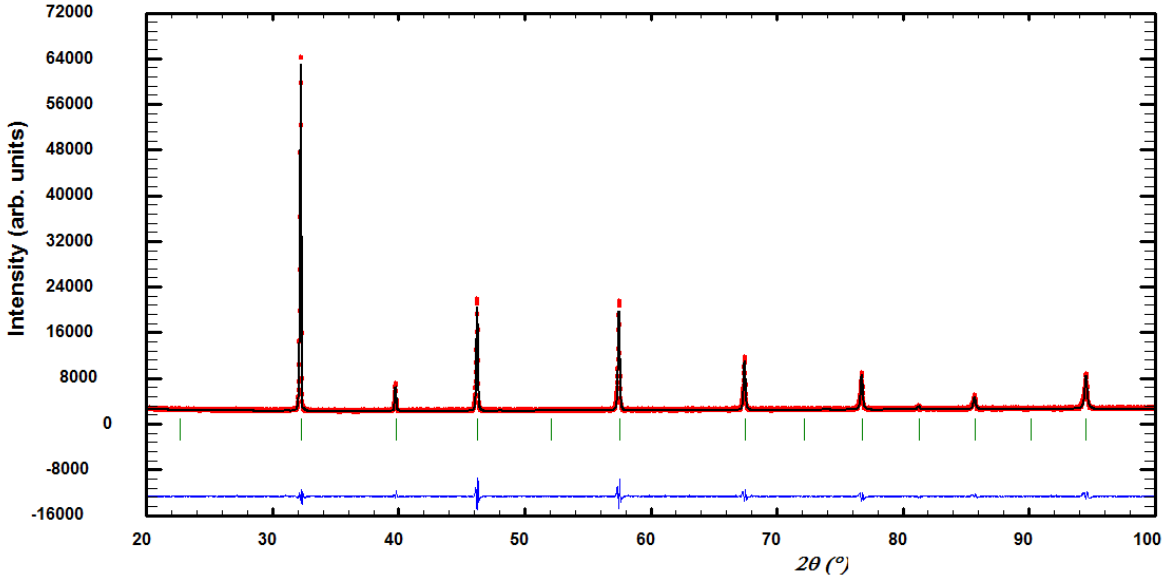

Figure SI 3. Experimental (red circles) and calculated (black continuous line) XRD patterns (and their difference, blue line at the bottom) for $Sr_2CoNb_{0.80}Ti_{0.20}O_6$.

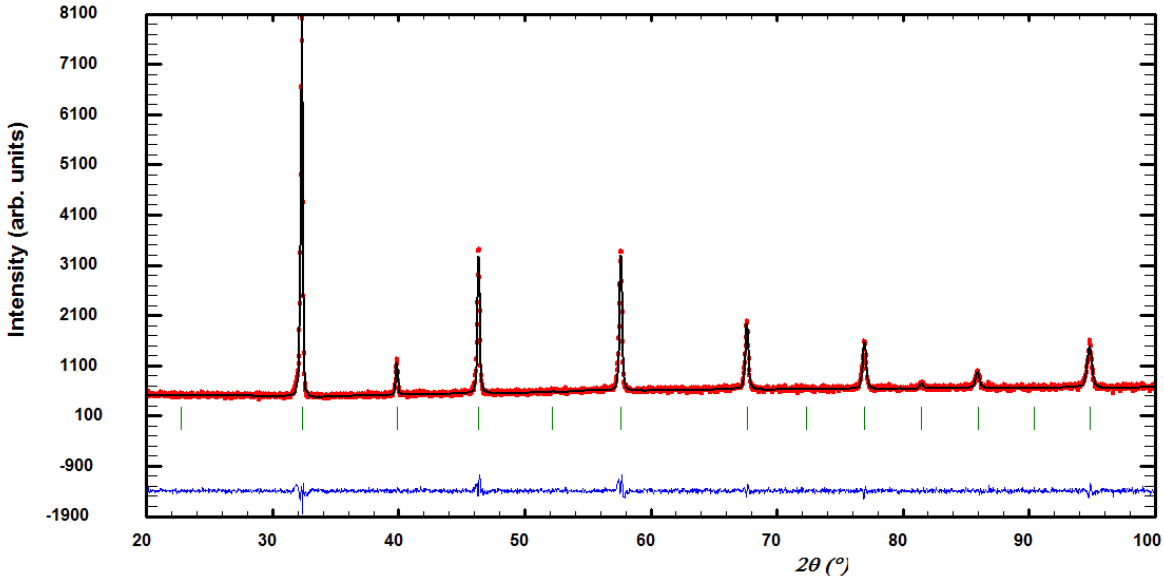

Figure SI 4. Experimental (red circles) and calculated (black continuous line) XRD patterns (and their difference, blue line at the bottom) for $Sr_2CoNb_{0.70}Ti_{0.30}O_6$.

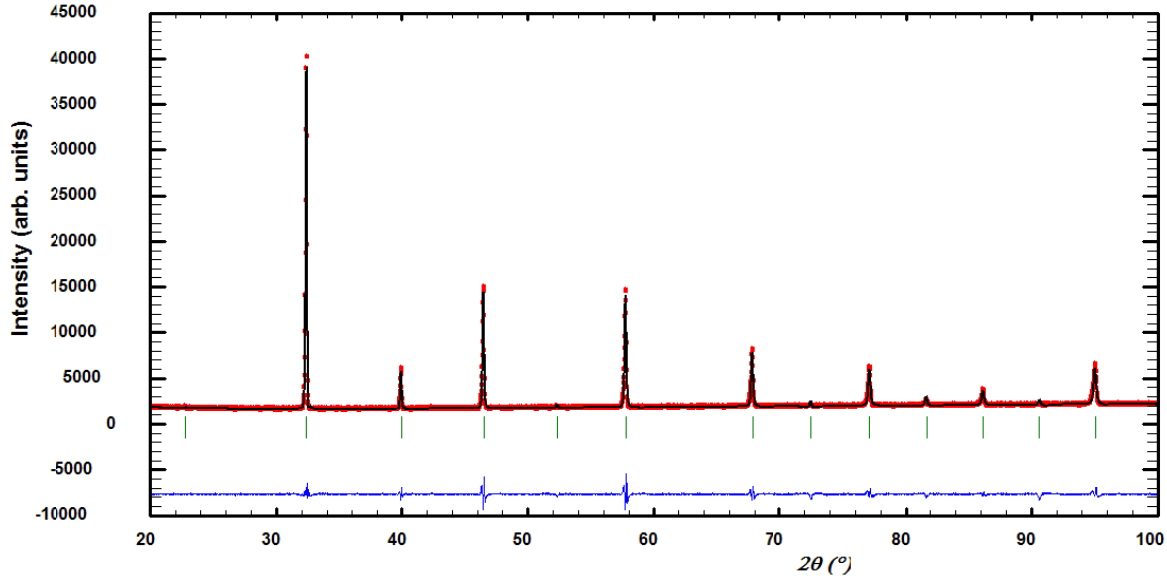

Figure SI 5. Experimental (red circles) and calculated (black continuous line) XRD patterns (and their difference, blue line at the bottom) for $Sr_2CoNb_{0.50}Ti_{0.50}O_6$.

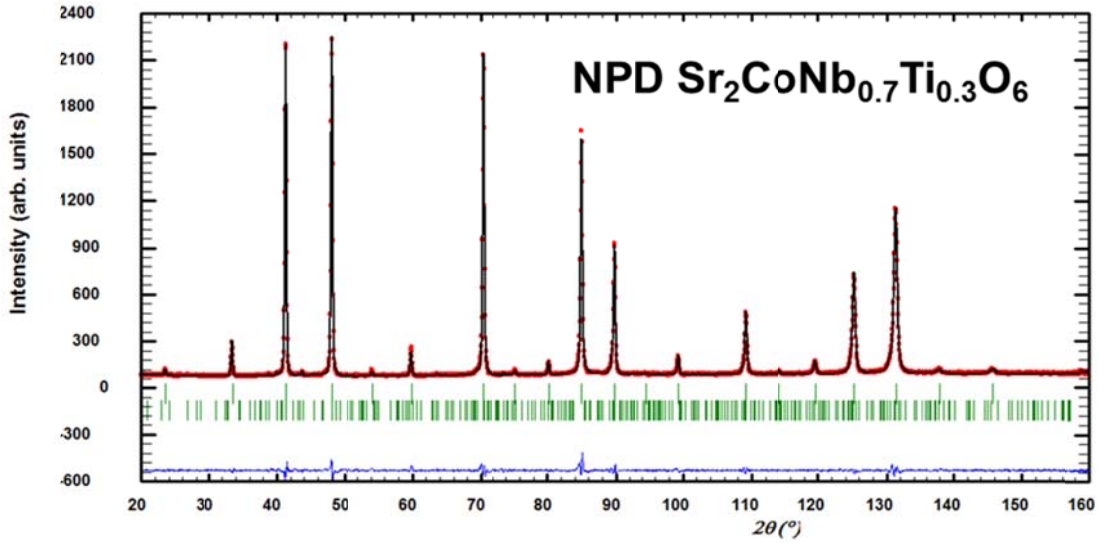

Figure SI 6. Experimental (red circles) and calculated (black continuous line) NPD patterns (and their difference, blue line at the bottom) for $Sr_2CoNb_{0.7}Ti_{0.3}O_6$. The first row of vertical bars indicates the positions of Bragg peaks for this material; the second row corresponds to a small amount (c.a. 1% weight) of $Sr_7Nb_6O_{21}$.

HREM of $Sr_2CoNb_{1-x}Ti_xO_6$ with x = 0.0

HREM taken along different zone axes were obtained to fully reconstruct the reciprocal/real space. The most relevant reciprocal zone axes corresponding to [001], [111] are given as SI2 and SI3, respectively; whereas that along [1-10] is presented in the manuscript as Figure 2. For the parent non-substituted material, images along [001] and [111] zone axes show an apparently ordered material with d-spacing corresponding to a simple perovskite structure, FT (inset of Fig. SI2 and SI3) reveals that the whole crystals seem to be homogeneous and only maxima corresponding to the simple cubic perovskite were observed.

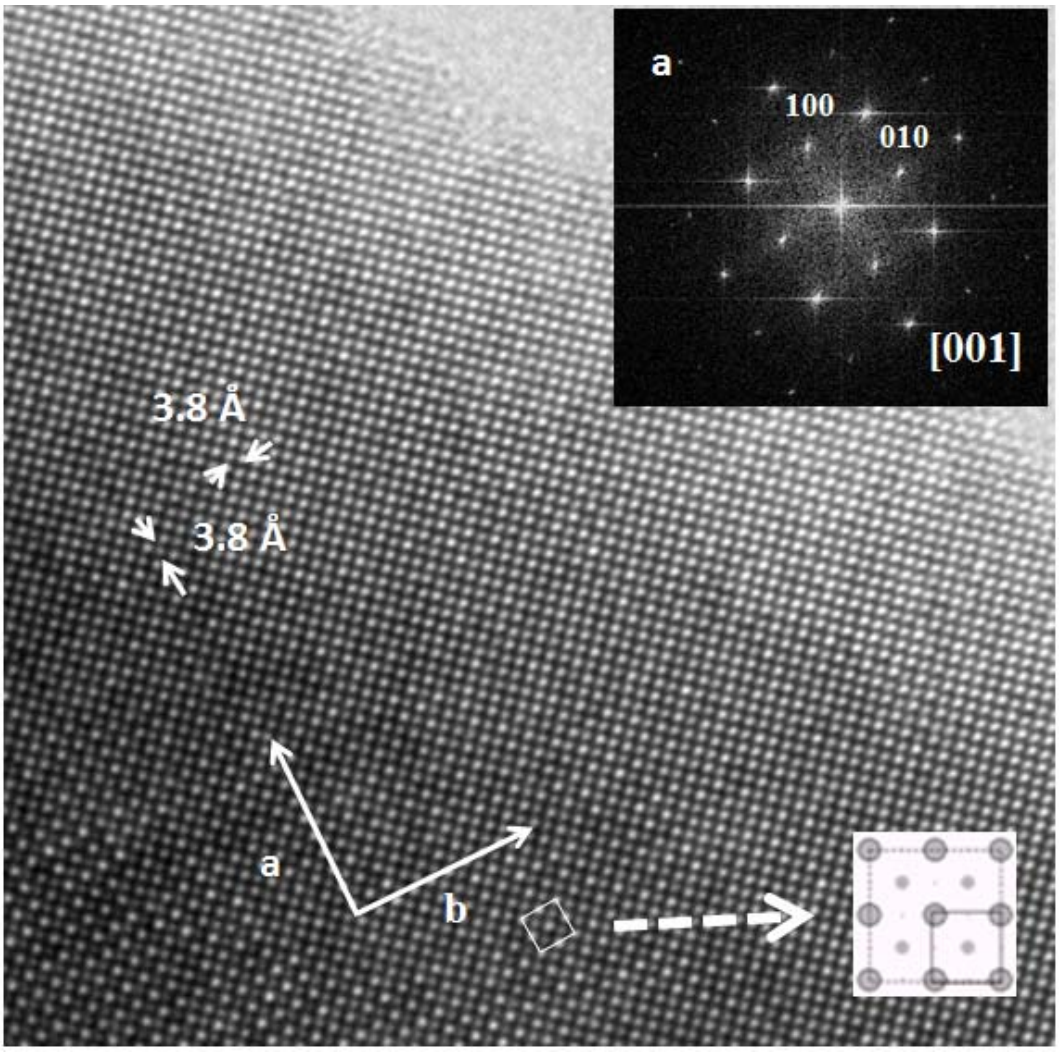

Figure SI 7. HREM image of Sr_2CoNbO_6 along [001]p. (a) Optical FT.

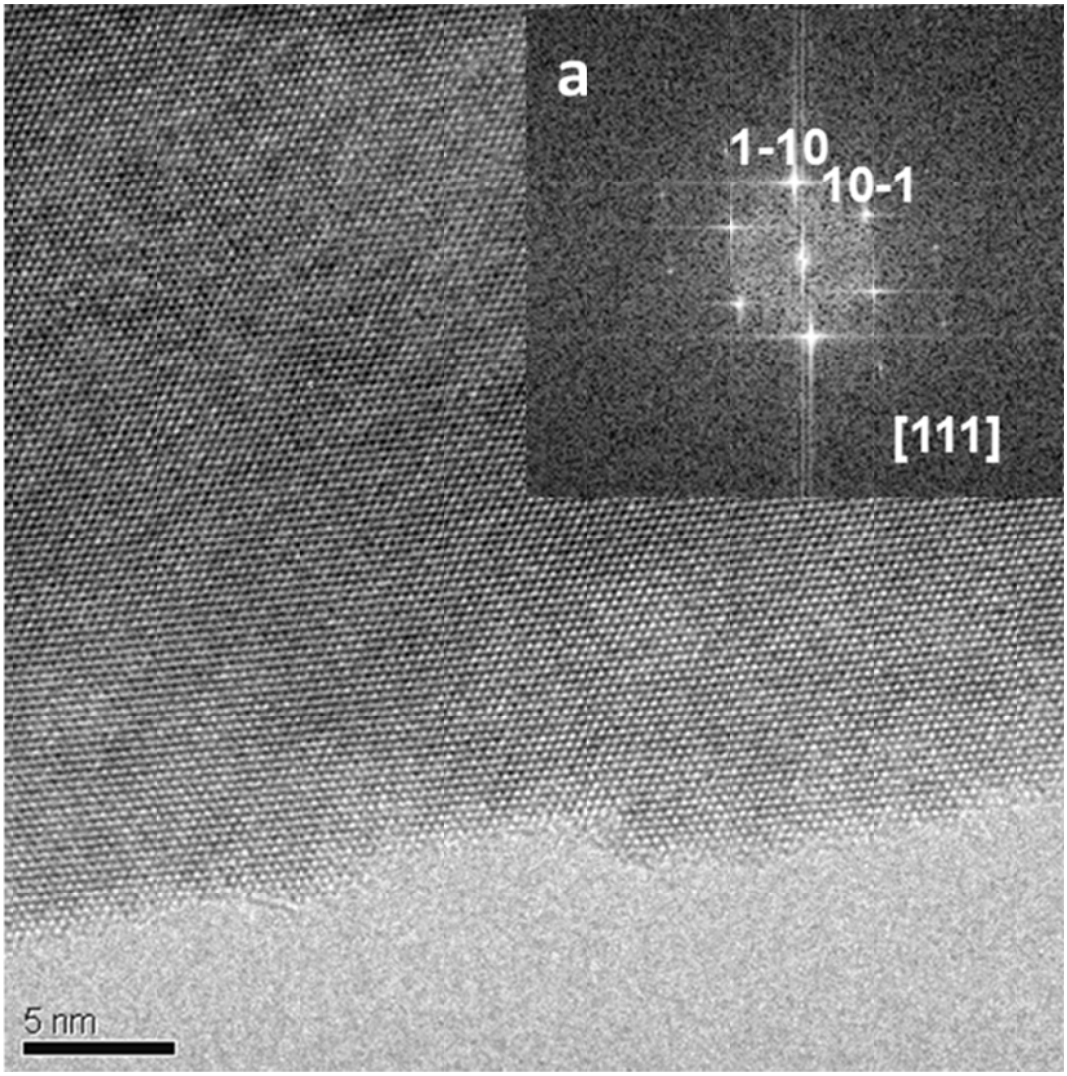

Figure SI 8. HREM image of Sr_2CoNbO_6 along [111]p. (a) Optical FT.

HREM **of $Sr_2CoNb_{1-x}Ti_xO_6$ with x = 0.1**

HREM taken along different zone axes were obtained to fully reconstruct the reciprocal/real space. The most relevant reciprocal zone axes corresponding to [001], [111] and [-110] are given as SI4, SI5 and SI6, respectively. As for the parent non-substituted material, Images along [001] and [111] zone axes show an apparently ordered material with d-spacing corresponding to a simple perovskite structure, FFT (inset of Fig. SI4 and SI5) reveals that the whole crystals seem to be homogeneous and only maxima corresponding to the simple cubic perovskite were observed.

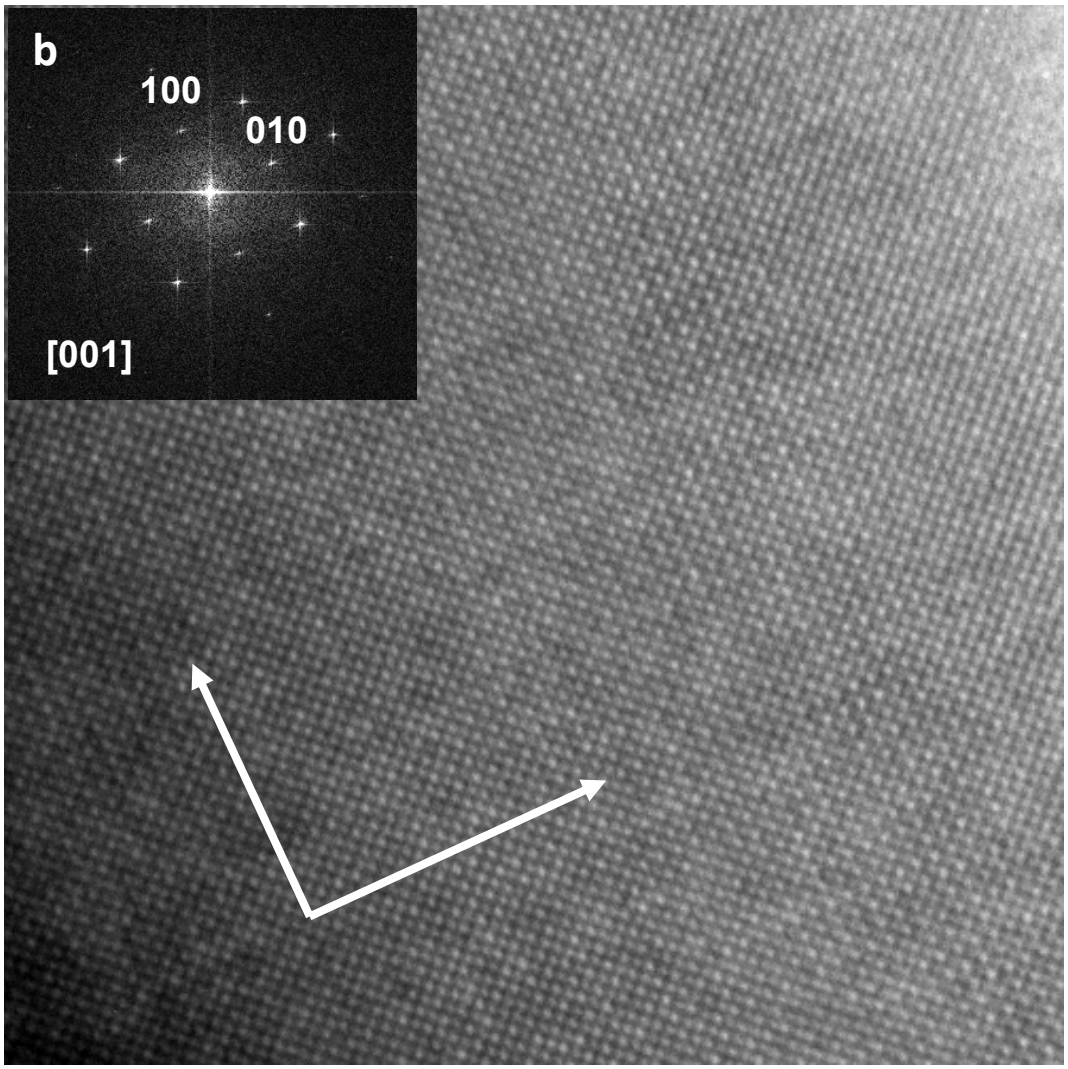

Figure SI 9. HREM image of $Sr_2CoNb_{0.9}Ti_{0.1}O_6$ along [001]p. (a) Optical FT.

SUPPLEMENTARY INFORMATION FOR

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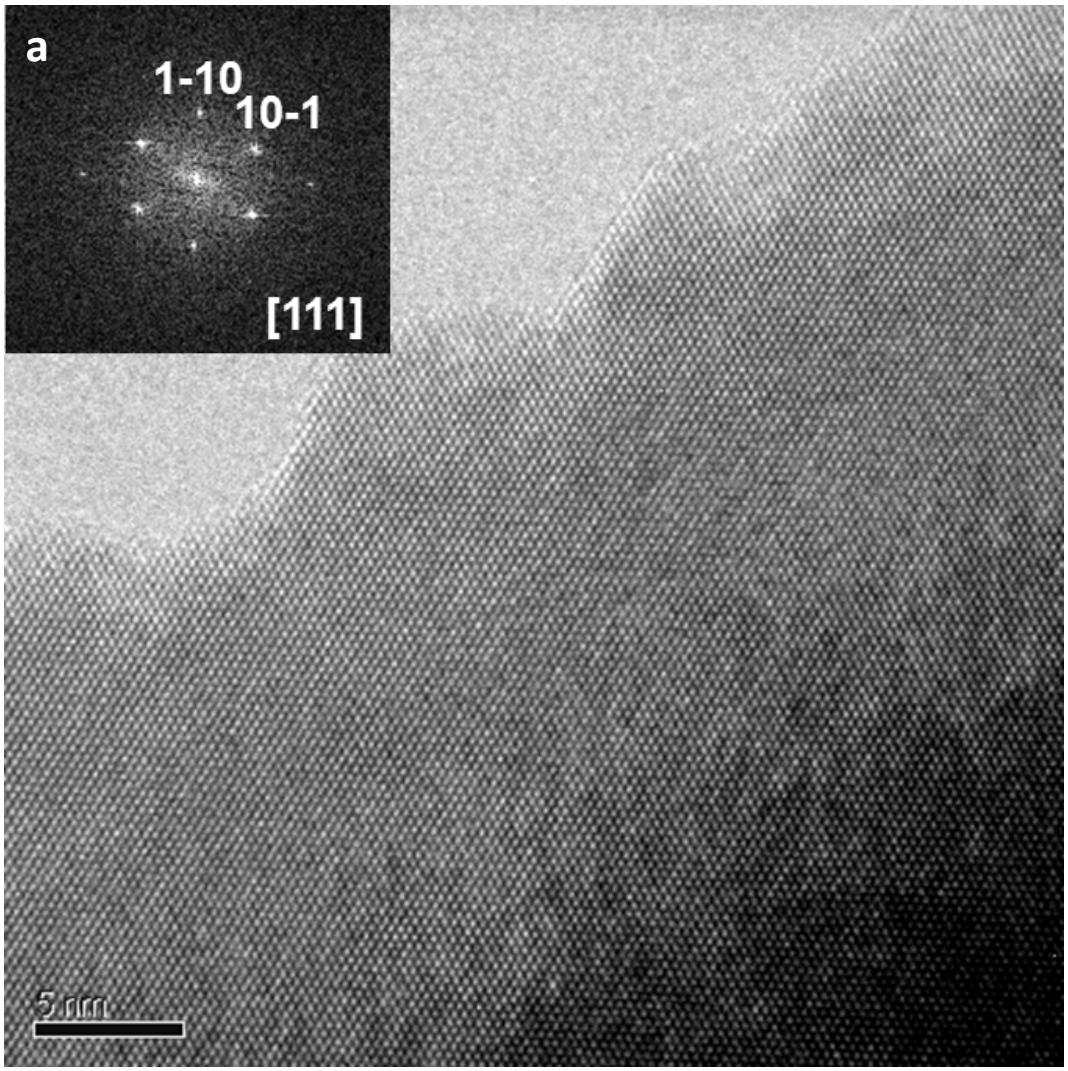

Figure SI 10. HREM image of $Sr_2CoNb_{0.9}Ti_{0.1}O_6$ along [111]p. (a) Optical FT.

As for the parent compound, HREM image along [-110]p revealed the existence of two kind of domains (simple cubic disordered and B-ions ordered) which intergrowth in a disordered fashion. In Fig.SI 7 FFTs of adjacent domains of both types are presented.

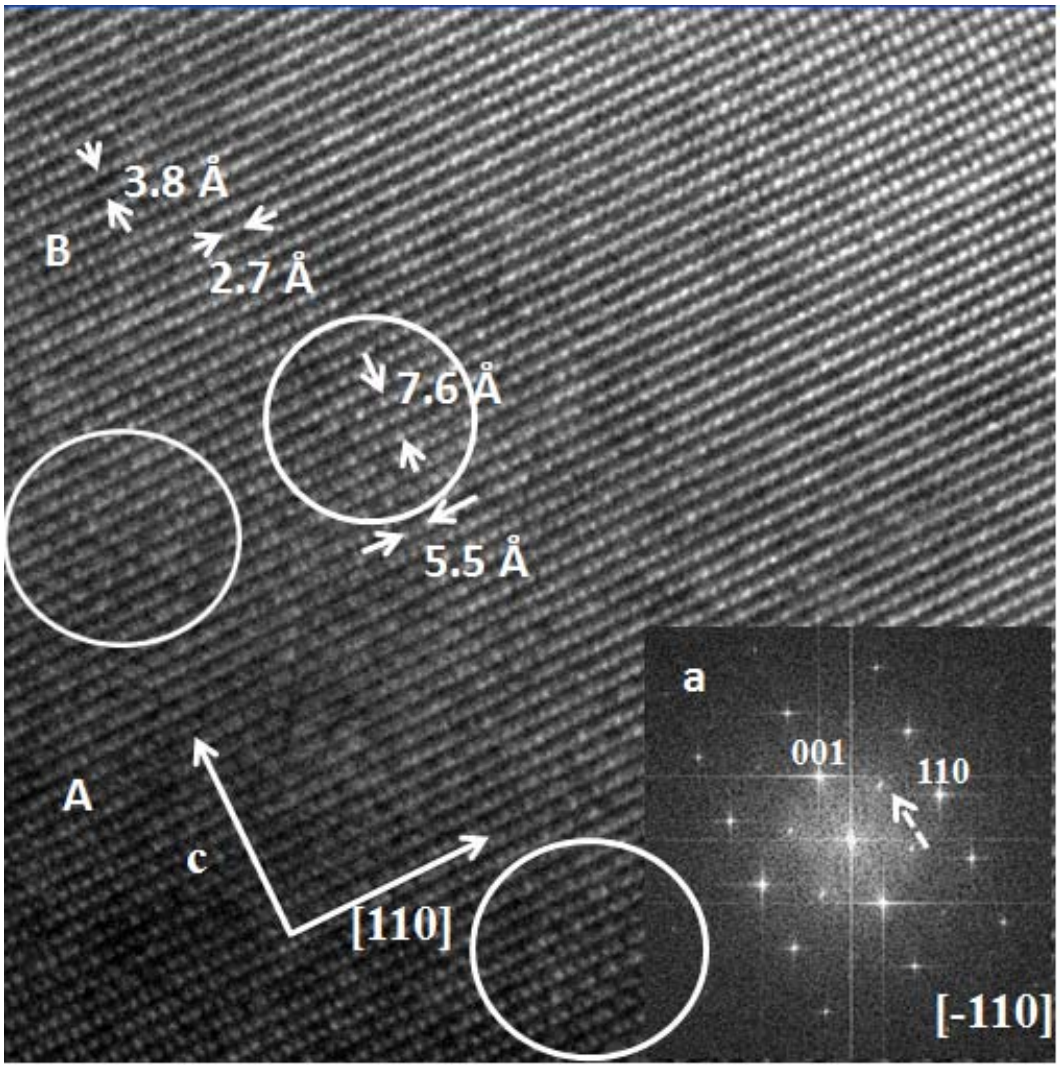

Figure SI 11. HREM image of $Sr_2CoNb_{0.9}Ti_{0.1}O_6$ along [110]p. (a) Optical FT.

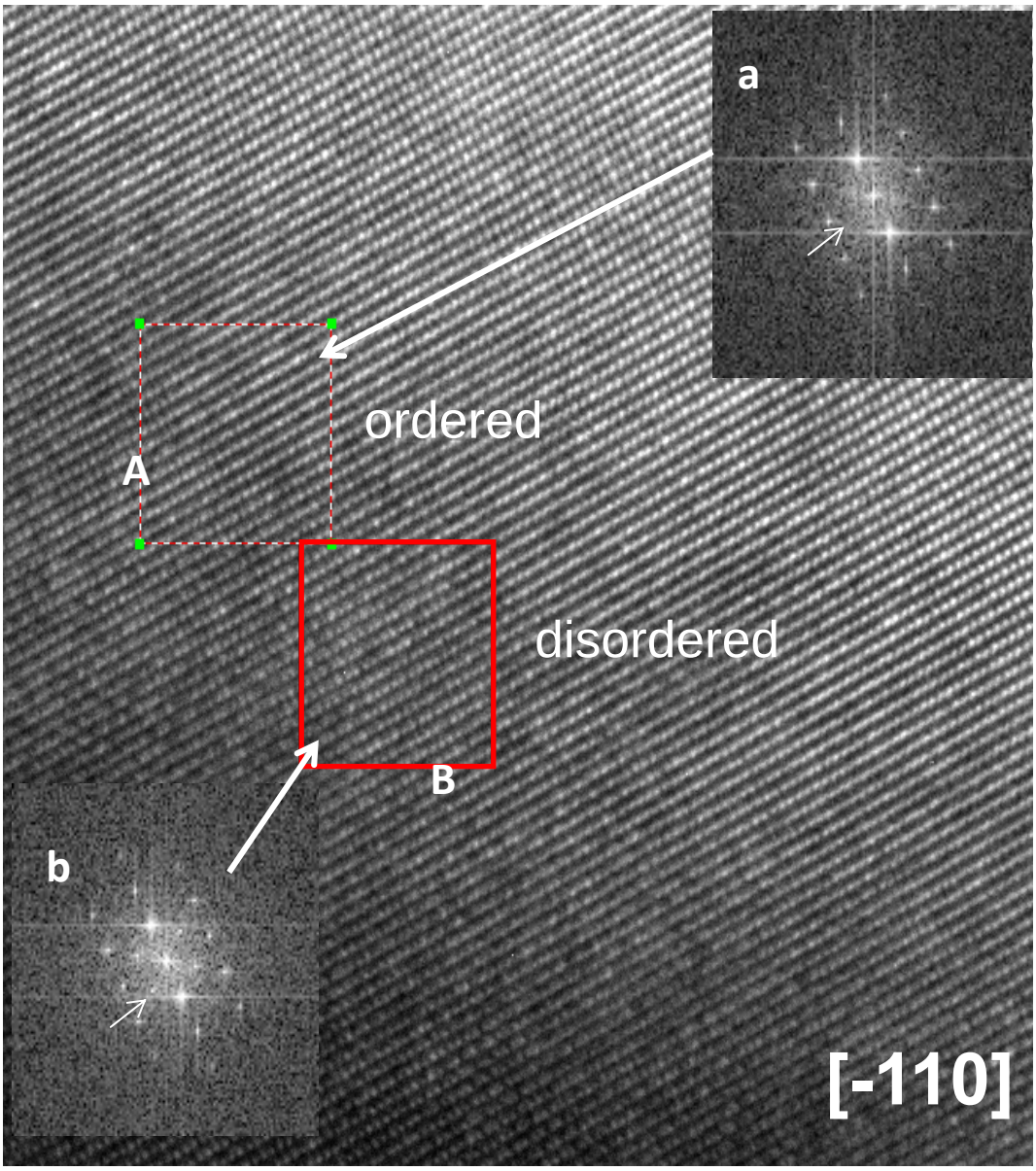

Figure SI 12. HREM image of $Sr_2CoNb_{0.9}Ti_{0.1}O_6$ along [110]p. (a) Optical FT corresponding to double-perovskite domains, the arrow indicates a superstructure spot, (b) Optical FT of a simple-cubic perovskite domain.

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HREM of $Sr_2CoNb_{1-x}Ti_xO_6$ with x = 0.2

HREM taken along different zone axes were obtained to fully reconstruct the reciprocal/real space. The most relevant reciprocal zone axes corresponding to [001], [111] and [110] are given as SI8, SI9 and SI10, respectively. As for the previous cases images along [001] and [111] zone axes show an apparently ordered material with d-spacing corresponding to a simple perovskite structure. FT (insets of Fig. SI8 and SI9) reveals that the whole crystals seem to be homogeneous and only maxima corresponding to the simple cubic perovskite are observed.

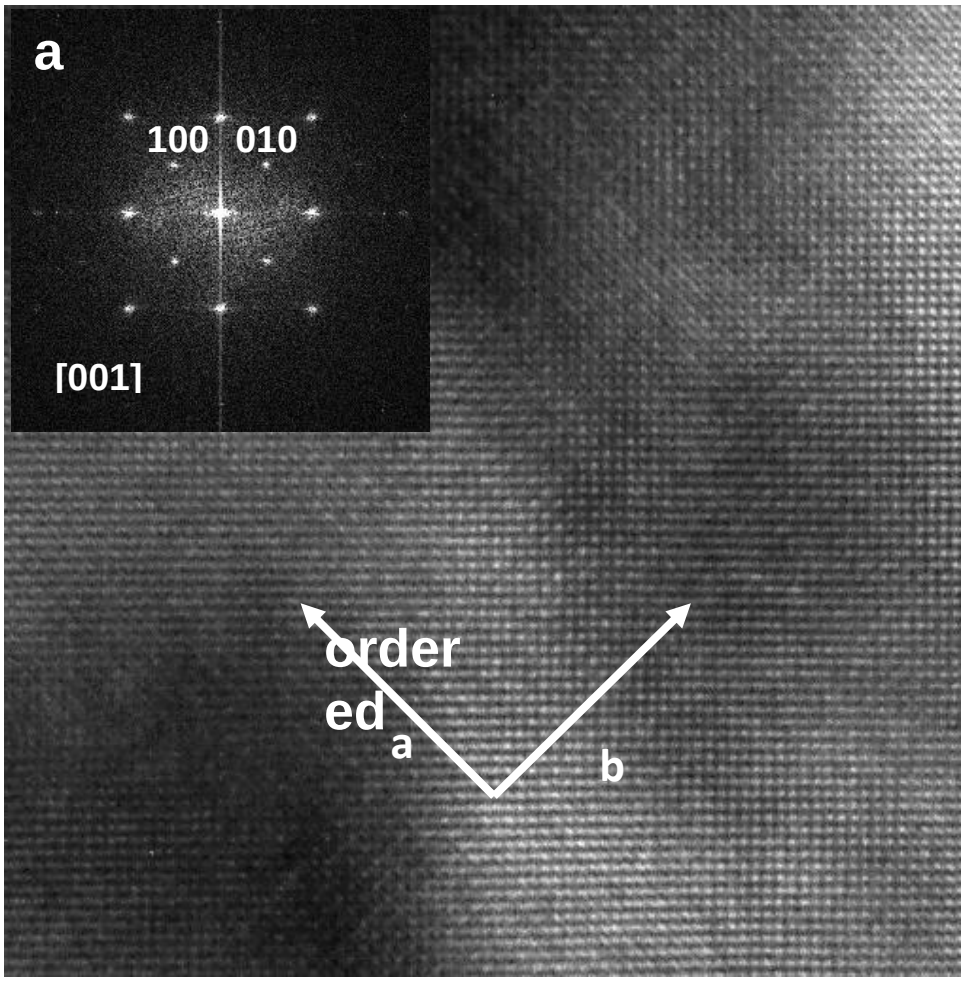

Figure SI 13. HREM image of $Sr_2CoNb_{0.8}Ti_{0.2}O_6$ along [001]p. (a) Optical FT.

SUPPLEMENTARY INFORMATION FOR

Complex magnetic behaviour of $Sr_2CoNb_{1-x}Ti_xO_6$ ($0 \leq x \leq 0.5$) as a result of a flexible microstructure

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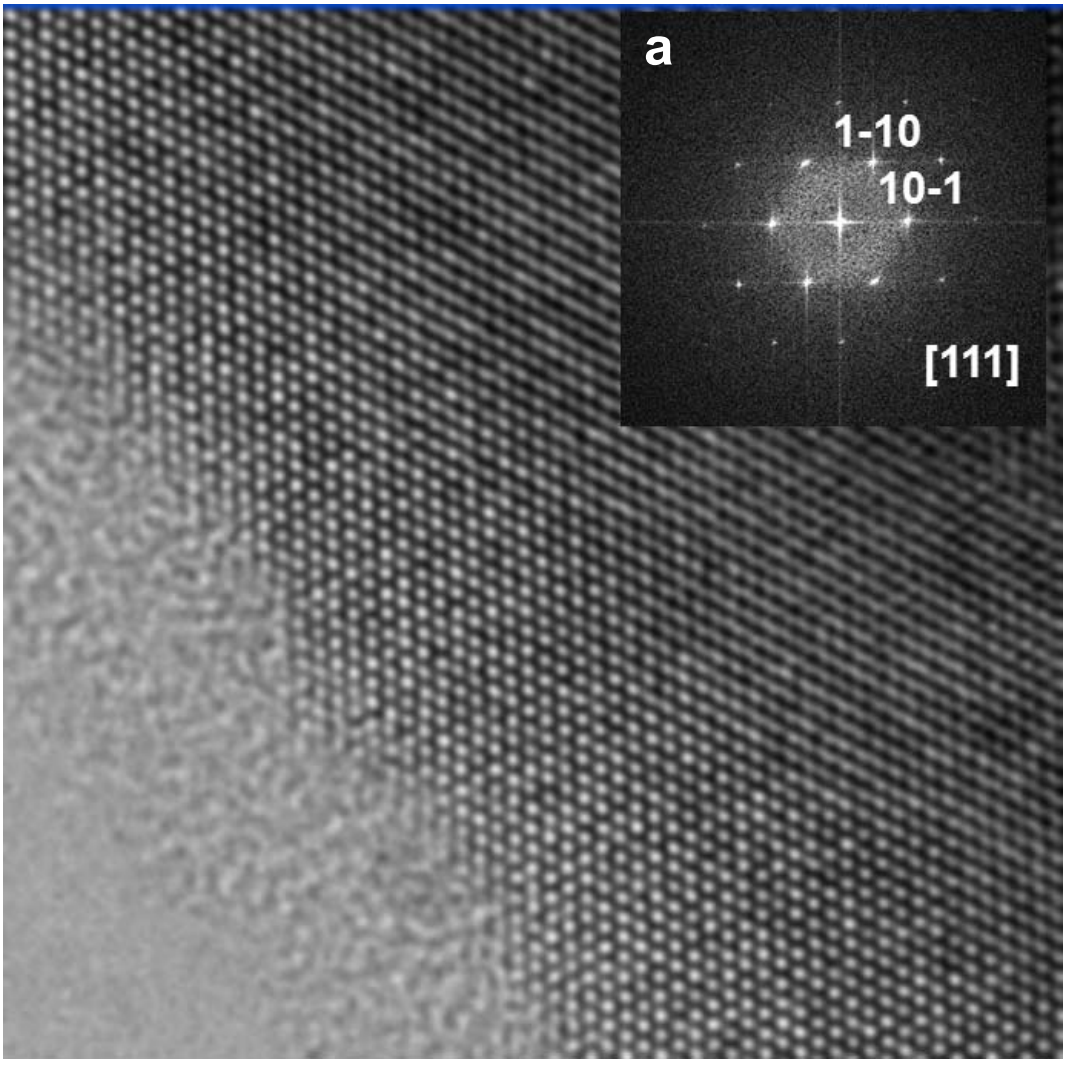

Figure SI 14. HREM image of $Sr_2CoNb_{0.8}Ti_{0.2}O_6$ along [111]p. (a) Optical FT.

HREM image along [110]p (Fig. SI 10) evidences the existence of small domains of double-ordered perovskite (indicated as A) which intergrowths on a matrix of a simple-

cubic-disordered perovskite (indicated as B). The size and number of ordered domains decrease in comparison to materials with lower Ti-contents. FT of both types of domains (Fig. SI 11) confirm the ordered (A) and disordered (B) structures.

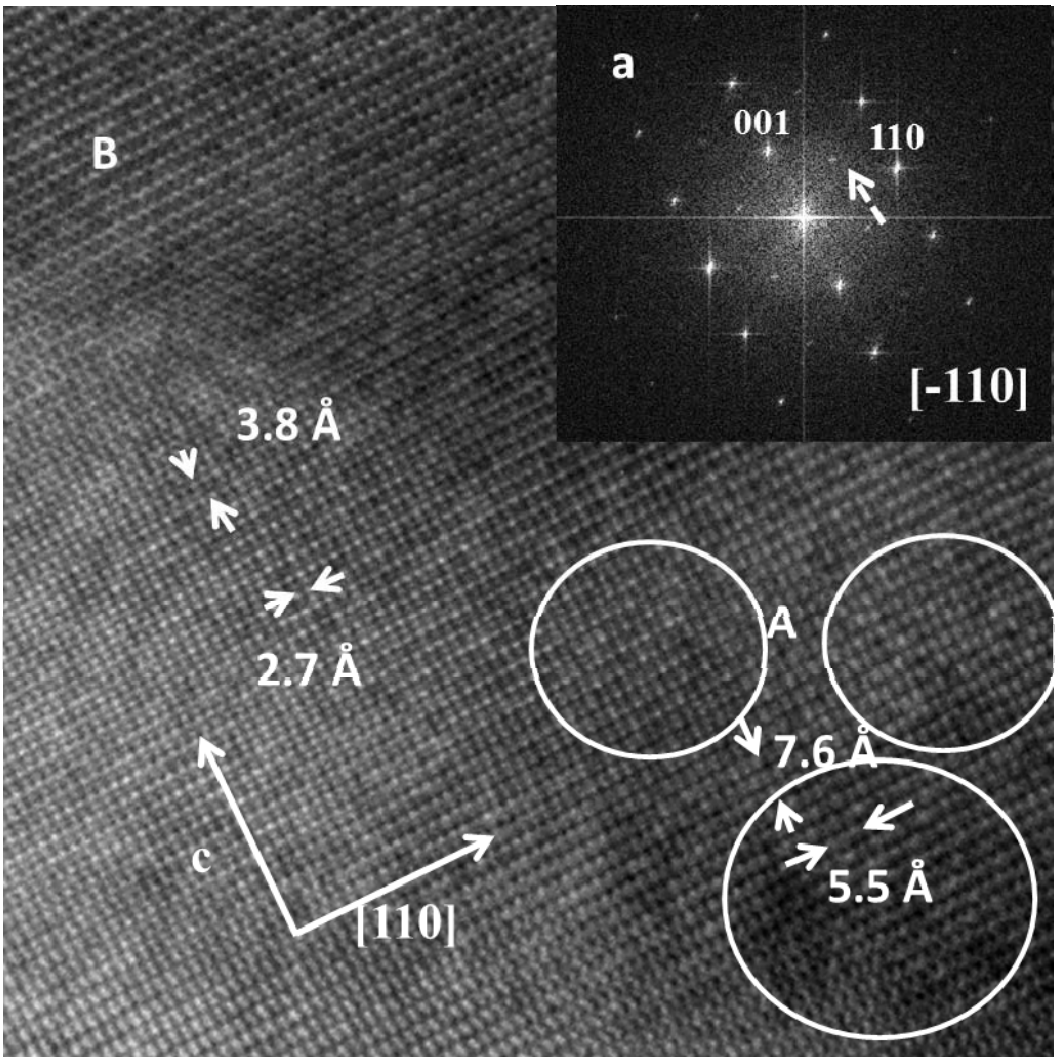

Figure SI 15. HREM image of $Sr_2CoNb_{0.8}Ti_{0.2}O_6$ along [110]p. (a) Optical FT corresponding to the superposition of ordered (A) where the arrow indicates a superstructure spot and disordered (B) domains.

SUPPLEMENTARY INFORMATION FOR

Complex magnetic behaviour of $Sr_2CoNb_{1-x}Ti_xO_6$ ($0 \leq x \leq 0.5$) as a result of a flexible microstructure

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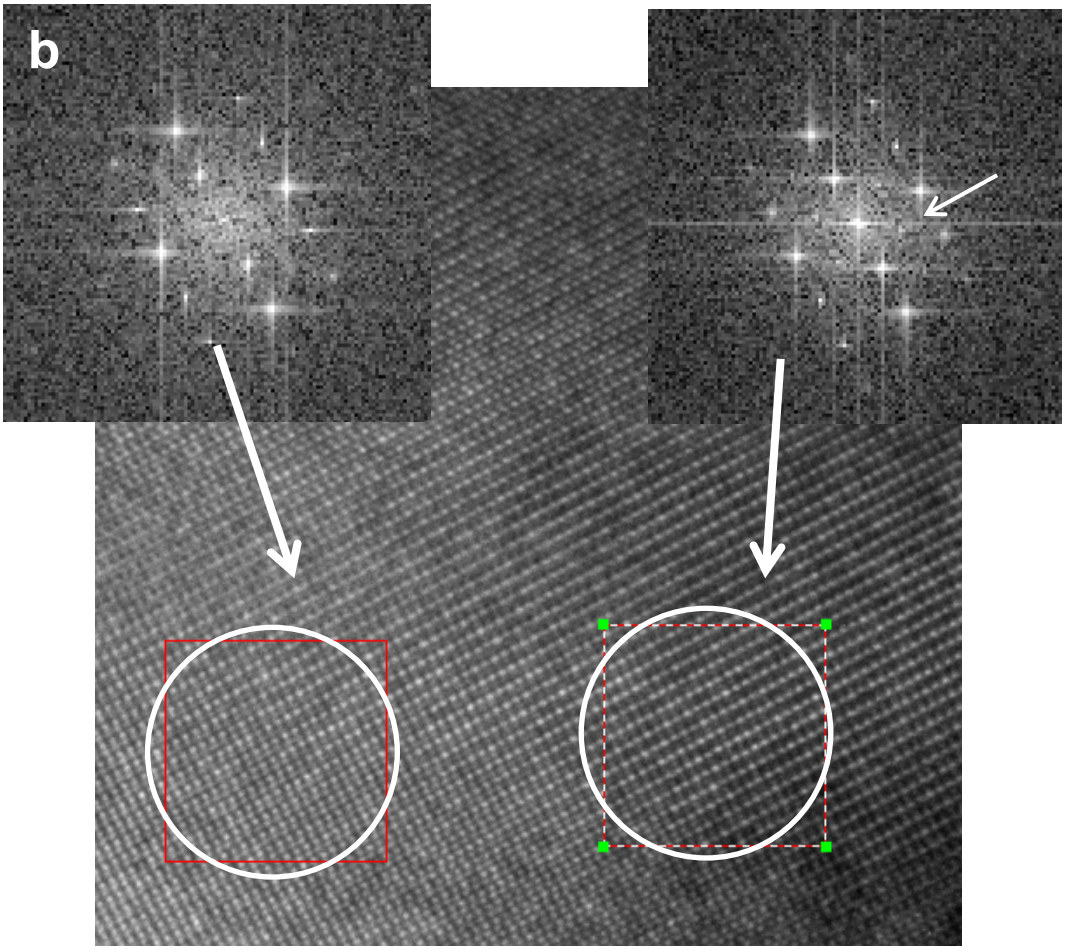


Figure SI 16. HREM image of $Sr_2CoNb_{0.8}Ti_{0.2}O_6$ along [110]p. (a) Optical FT corresponding to double-perovskite domains (A) where superstructure spot is indicated by an arrow, (b) Optical FT of a simple-cubic perovskite domains (B).

HREM of $Sr_2CoNb_{1-x}Ti_xO_6$ with x = 0.3

For $Sr_2CoNb_{0.7}Ti_{0.3}O_6$, the electron microscopy study (Figs. SI 12 and SI 13) shows that some small domains of ordered perovskite structure (few unit cells areas) still remain along the simple-perovskite matrix.

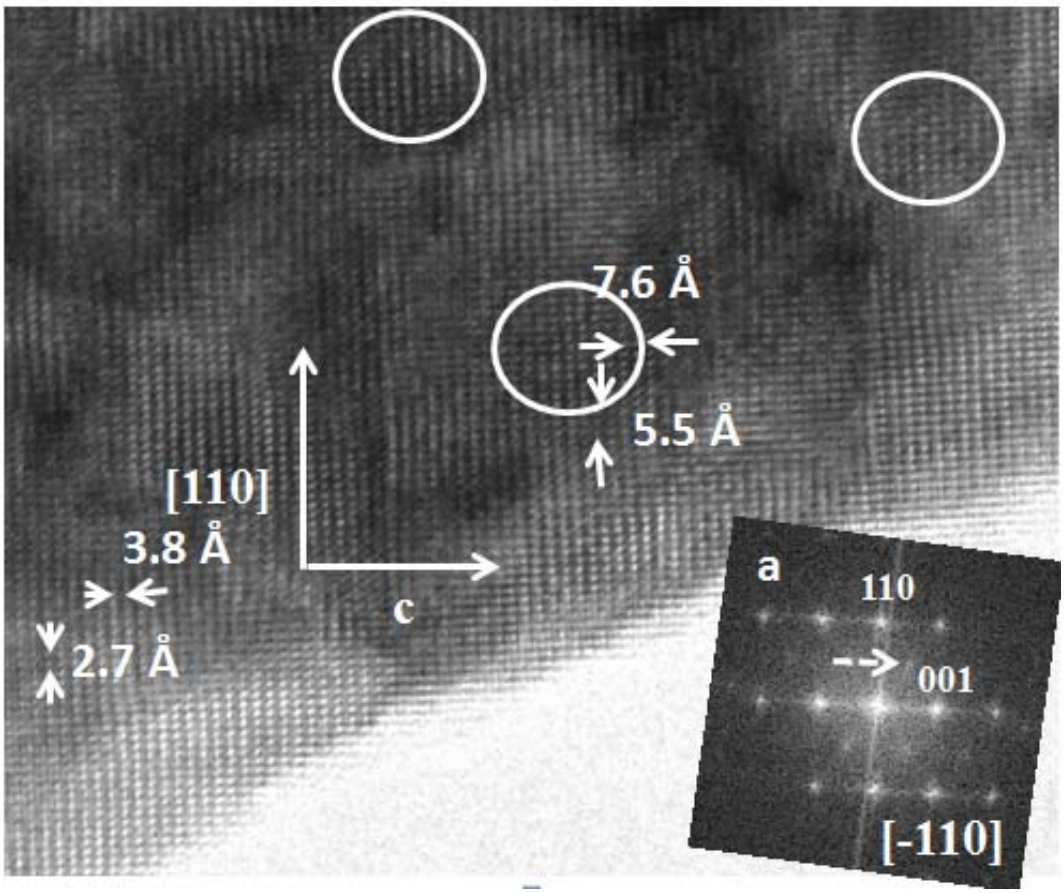


Figure SI 17. HREM image of $Sr_2CoNb_{0.7}Ti_{0.3}O_6$ along [1-10]p. (a) Optical FT, the arrow indicates a superstructure spot.

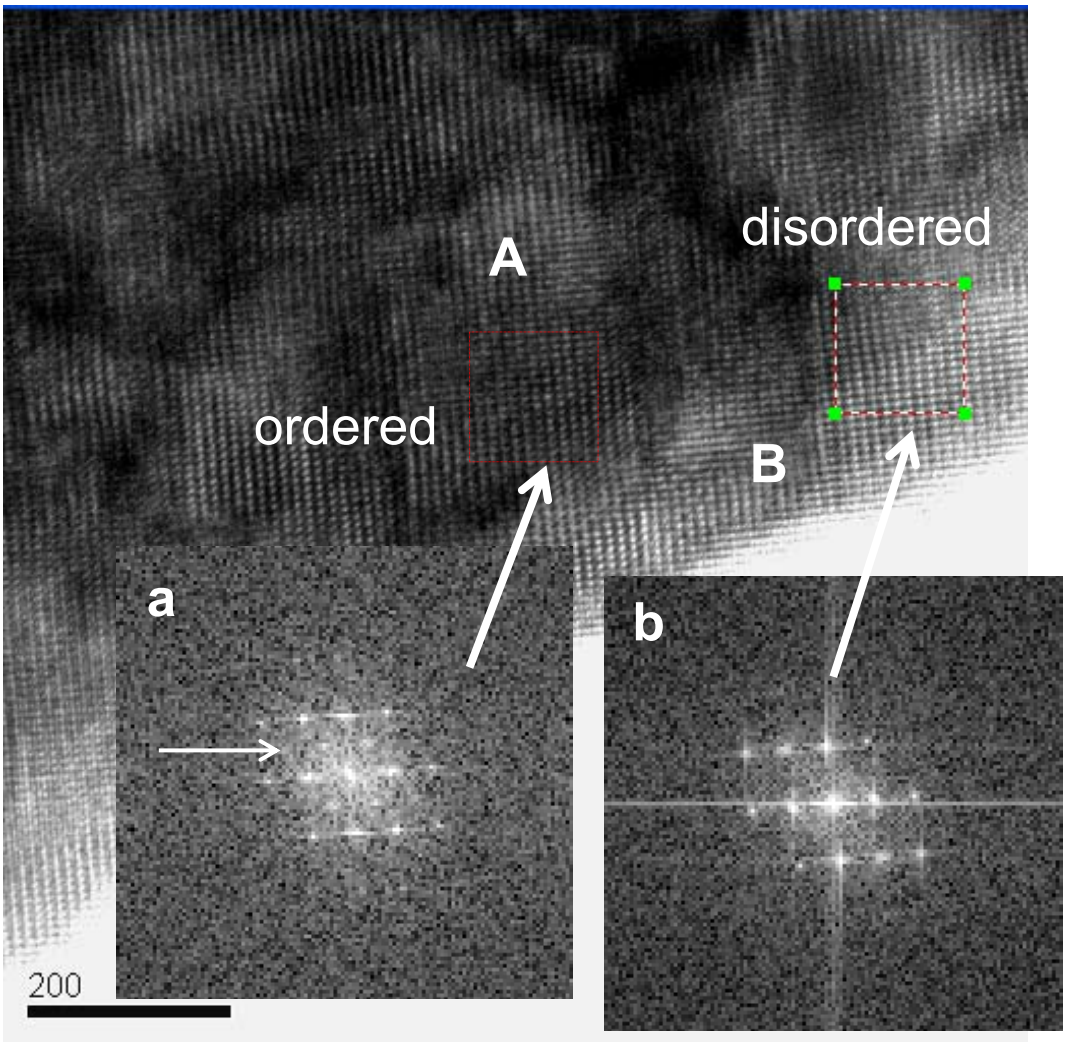


Figure SI 18. HREM image of $Sr_2CoNb_{0.7}Ti_{0.3}O_6$ along [110]p. (a) Optical FT corresponding to double-perovskite domains (A) the superstructure spot is indicated by an arrow, (b) Optical FT of a simple-cubic perovskite domains (B).

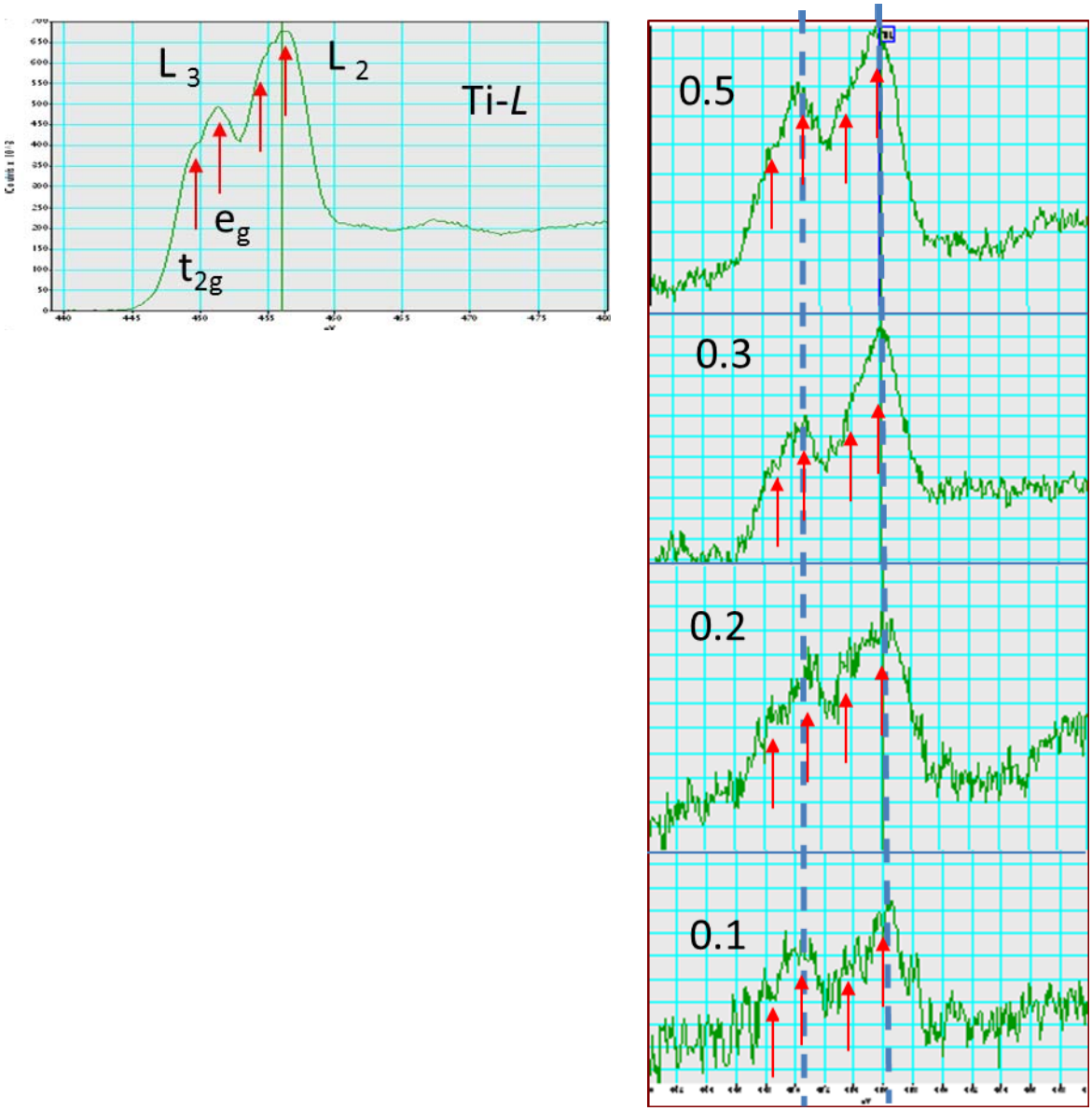


Figure SI 19. EELS spectra (right) of the Ti(L_2,L_3) edges for 0.1 ≤ x ≤0.5 of $Sr_2CoNb_{1-x}Ti_xO_6$ compounds. The reference Ti^{4+} spectrum (left) was acquired on rutile TiO_2 compound as internal standard.

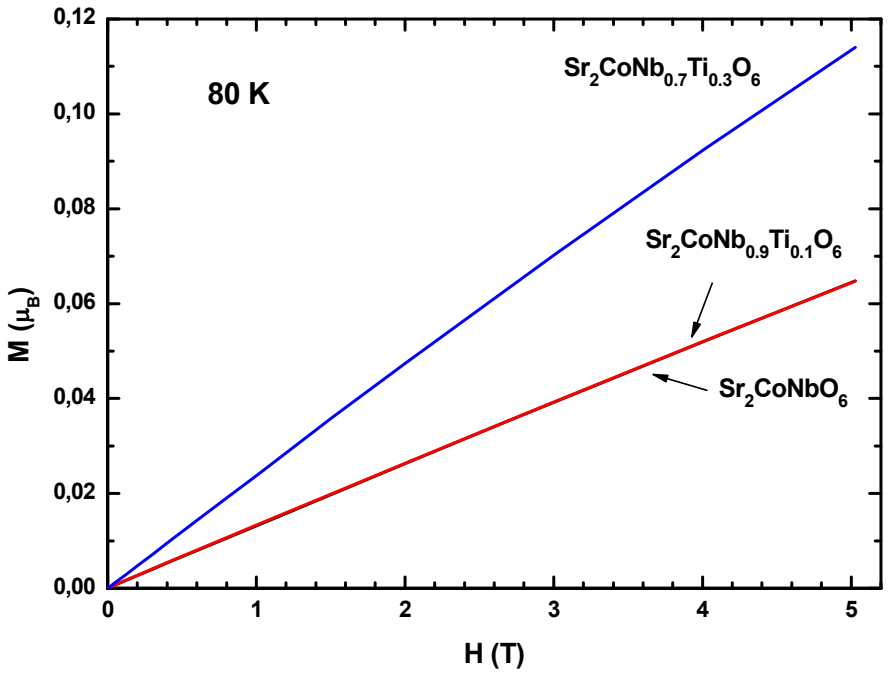
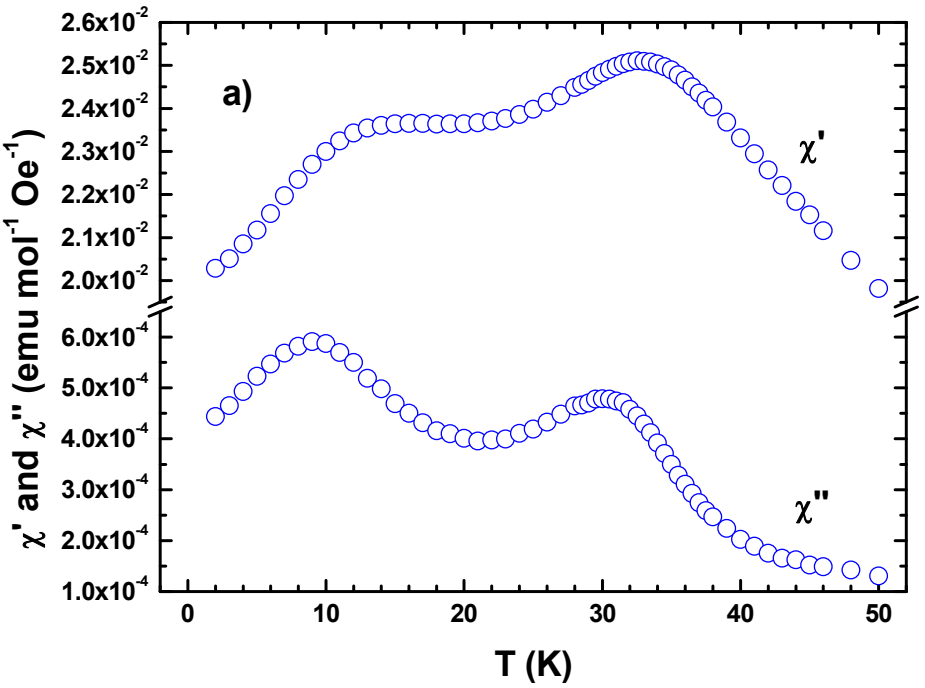
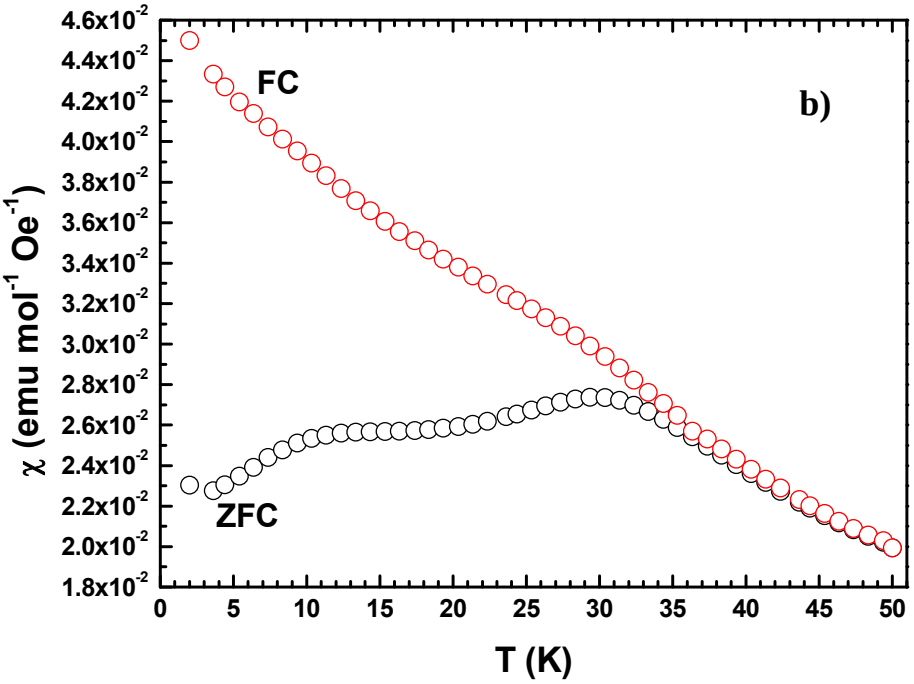


Figure SI 20.: Applied magnetic field dependence of magnetization at 80 K for Sr_2CoNbO_6, $Sr_2CoNb_{0.9}Ti_{0.1}O_6$ and $Sr_2CoNb_{0.7}Ti_{0.3}O_6$ oxides.


a)
χ'
χ''
χ' and χ'' (emu mol-1 Oe-1)
T (K)
2.6x10-2
2.5x10-2
2.4x10-2
2.3x10-2
2.2x10-2
2.1x10-2
2.0x10-2
6.0x10-4
5.0x10-4
4.0x10-4
3.0x10-4
2.0x10-4
1.0x10-4
0
10
20
30
40
50


FC
ZFC
b)
χ (emu mol-1 Oe-1)
T (K)
4.6x10-2
4.4x10-2
4.2x10-2
4.0x10-2
3.8x10-2
3.6x10-2
3.4x10-2
3.2x10-2
3.0x10-2
2.8x10-2
2.6x10-2
2.4x10-2
2.2x10-2
2.0x10-2
1.8x10-2
0
5
10
15
20
25
30
35
40
45
50

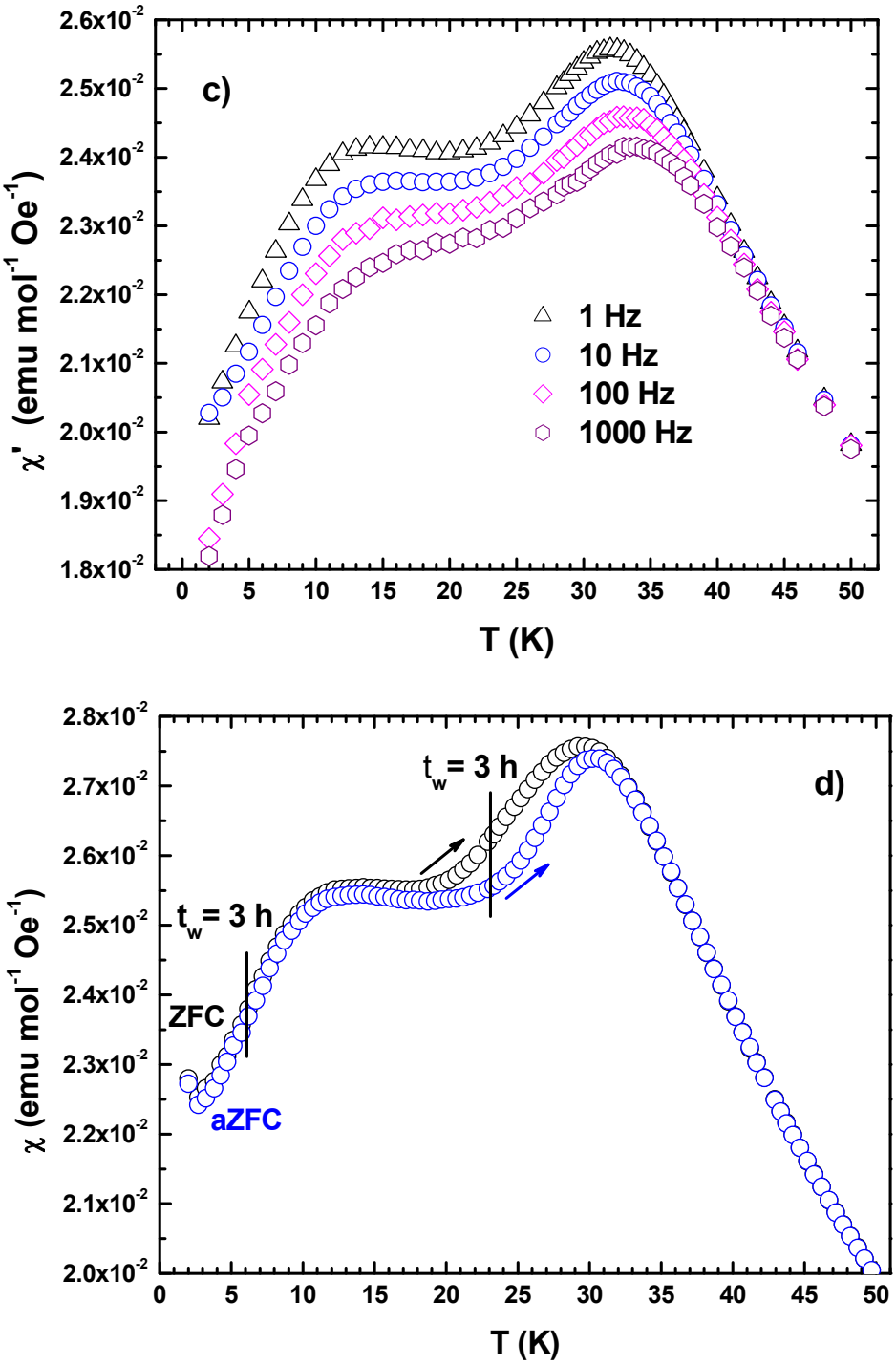


Figure SI 21. For $Sr_2CoNb_{0.7}Ti_{0.3}O_6$ oxide: a) Temperature dependence of real and imaginary parts of ac-χ measured at a driving field of 0.35 mT and 10 Hz (H_{dc}= 0 T). b) Temperature dependence of dc-χ measured in an applied magnetic field of 10 mT following the ZFC and FC protocols. c) Frequency dependence of χ' measured at a driving field of 0.35 mT with frequencies 1, 10, 100 and 1000 Hz. d) Temperature dependence of aged χ^{ZFC} (obtained waiting 3 hours at 23 and 6 K, consecutively) and not aged (normal) χ^{ZFC} measured in an applied magnetic field of 5 mT.

Table SI 1: Structural parameters for $Sr_2CoNb_{1-x}Ti_xO_6$ (x = 0.30, 0.50) compounds obtained from NPD data.

	[b] $Sr_2CoNb_{0.70}Ti_{0.30}O_6$	[c] $Sr_2CoNb_{0.50}Ti_{0.50}O_6$
[a] **S.G. Pm-3m**		
***a* (Å)**	3.90322(2)	3.89620(2)
Sr position	1b	1b
Occ Sr	2	2
U*100 (Å^2)	0.70(2)	0.76(2)
B position	1a	1a
Occ Co/Nb//Ti	1/0.700(2)/0.289(2)	1/0.499(2)/0.501(2)
U*100 (Å^2)	0.79(2)	0.90(2)
O(1) position	3d	3d
Occ	6.003(2)	5.942(2)
U*100 (Å^2)	0.95(3)	1.01(3)

[a] 1a (000), 1b (½ ½ ½), 3d (½ 0 0)

[b] χ^2= 3.66, R_{wp}= 3.96%, R_{exp}= 2.07%, R_B= 1.09%, Composition: $Sr_2CoNb_{0.700(2)}Ti_{0.289(2)}O_{6.003(2)}$

[c] χ^2= 3.51, R_{wp}= 3.90%, R_{exp}= 2.08%, R_B= 1.37%, Composition: $Sr_2CoNb_{0.499(2)}Ti_{0.501(2)}O_{5.942(2)}$