

Figure SI 1. Experimental (red points), calculated (black solid line) and their difference (blue line at bottom) XRD patterns for $\text{Sr}_2\text{EuNb}_{1-x}\text{Ti}_x\text{O}_{6-x/2}$ with $x=0.25$; the vertical bars indicate the positions of Bragg peaks for this phase (upper bars) and for $\text{Sr}_3\text{Ti}_2\text{O}_7$ (lower bars). In the zoomed region weak peaks of an unidentified impurity phase are indicated by asterisks.

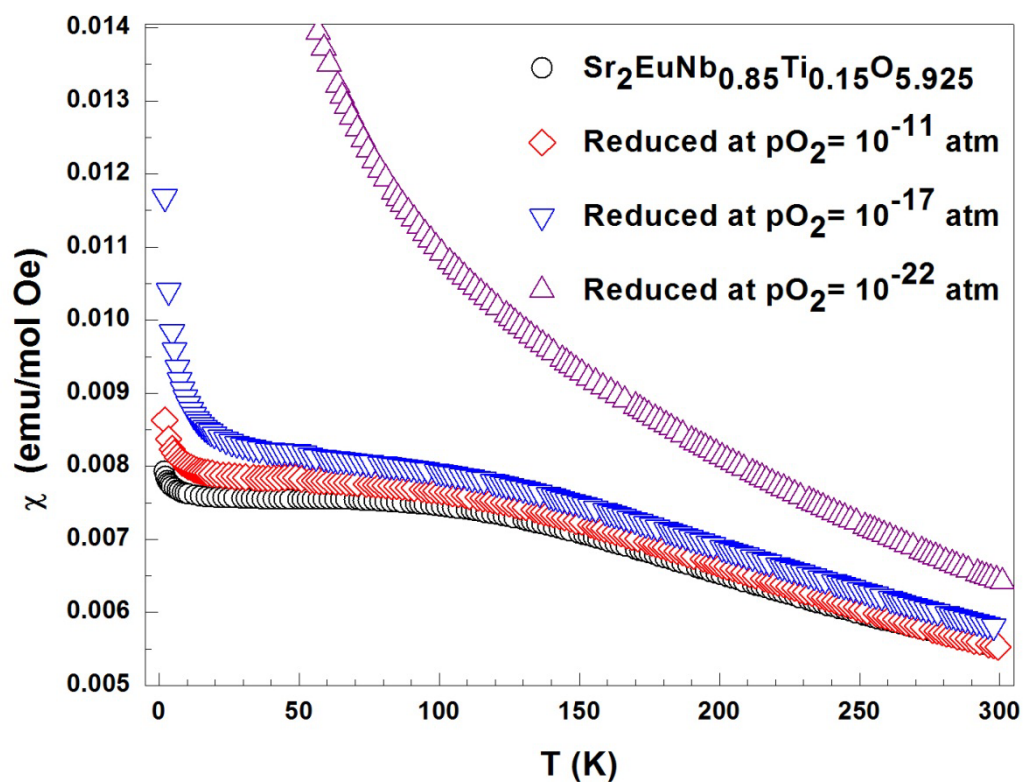


Figure SI 2. Magnetic susceptibility as a function of temperature for as-prepared $\text{Sr}_2\text{EuNb}_{0.85}\text{Ti}_{0.15}\text{O}_{5.925}$ and samples reduced under different $p\text{O}_2$ conditions. As the $p\text{O}_2$ decreases the magnetic susceptibility increases due to the consequent higher Eu^{2+} content in the material.

Table SI 1: Structural parameters for Sr₂EuNb_{1-x}Ti_xO_{6-x/2} (0 ≤ x ≤ 0.20) compounds obtained from XRD.

	^a Sr ₂ EuNbO ₆	^b Sr ₂ EuNb _{0.95} Ti _{0.05} O ₆	^c Sr ₂ EuNb _{0.90} Ti _{0.10} O ₆	^d Sr ₂ EuNb _{0.85} Ti _{0.15} O ₆	^e Sr ₂ EuNb _{0.80} Ti _{0.20} O ₆
a (Å)	5.8438(2)	5.84134(5)	5.8385(3)	5.8383(4)	5.8393(1)
b (Å)	5.9255(2)	5.91717(5)	5.9028(3)	5.8973(4)	5.8920(1)
c (Å)	8.3072(3)	8.30257(7)	8.2956(5)	8.2895(5)	8.2873(1)
β (deg)	90.216(2)	90.204(1)	90.198(4)	90.193(4)	90.183(1)
Sr position	4e	4e	4e	4e	4e
Occ Sr	1.00	1.00	1.00	1.00	1.00
x	-0.005(2)	-0.005(1)	-0.002(3)	-0.002(3)	0.007(2)
y	0.4654(6)	0.4657(4)	0.4701(6)	0.4712(8)	0.4736(6)
Z	0.2521(9)	0.2507(7)	0.251(1)	0.253(1)	0.249(1)
U*100 (Å²)	0.70(2)	0.21(2)	0.62(3)	0.76(4)	0.58(3)
Nb/Ti position	2b	2b	2b	2b	2b
Occ Nb/Ti	0.50/0.00	0.473(4)/0.027(4)	0.459(6)/0.041(6)	0.432(5)/0.068(5)	0.420(4)/0.080(4)
U*100 (Å²)	0.62(4)	0.35(3)	0.71(4)	0.68(5)	0.36(3)
Eu position	2a	2a	2a	2a	2a
Occ Eu	0.50	0.50	0.50	0.50	0.50
U*100 (Å²)	0.76(4)	0.28(3)	0.68(3)	0.73(4)	0.62(4)
O(1) position	4e	4e	4e	4e	4e
X	0.075(4)	0.095(3)	0.093(5)	0.131(6)	0.104(5)
y	0.0378(4)	0.025(3)	0.036(5)	0.070(5)	0.037(6)
Z	0.261(4)	0.260(3)	0.269(7)	0.261(7)	0.294(6)
Occ	1.00	1.00	1.00	1.00	1.00
U*100 (Å²)	0.76(3)	0.53(3)	0.82(5)	0.84(4)	0.72(4)
O(2) position	4e	4e	4e	4e	4e
X	0.235(6)	0.231(4)	0.236(7)	0.325(7)	0.222(5)
y	0.308(5)	0.312(3)	0.303(7)	0.308(7)	0.314(6)
Z	-0.034(5)	-0.035(4)	0.021(6)	0.024(6)	-0.033(5)
Occ	1.00	1.00	1.00	1.00	1.00
U*100 (Å²)	0.76(3)	0.53(3)	0.82(5)	0.84(4)	0.72(4)
O(3) position	4e	4e	4e	4e	4e
X	0.293(5)	0.291(4)	0.286(5)	0.245(7)	0.308(6)
y	0.757(5)	0.776(4)	0.773(6)	0.747(8)	0.793(6)
Z	-0.063(4)	-0.044(4)	-0.084(5)	-0.057(5)	-0.061(6)
Occ	1.00	1.00	1.00	1.00	1.00
U*100 (Å²)	0.76(3)	0.53(3)	0.82(5)	0.84(4)	0.72(4)

Space Group: P2₁/n (#14): 2a(000), 2b (00½), 4e (xyz),

^aχ²= 1.08, R_{wp}= 5.14%, R_{exp}= 4.96%, R_B= 3.10%, Composition: Sr₂EuNbTiO₆

^bχ²= 1.47, R_{wp}= 4.49%, R_{exp}= 3.70%, R_B= 5.10%, Composition: Sr₂EuNb_{0.95(2)}Ti_{0.05(2)}O₆

^cχ²= 1.11, R_{wp}= 6.33%, R_{exp}= 6.01%, R_B= 5.57%, Composition: Sr₂EuNb_{0.92(2)}Ti_{0.08(2)}O₆

^dχ²= 1.18, R_{wp}= 6.17%, R_{exp}= 5.71%, R_B= 5.68%, Composition: Sr₂EuNb_{0.86(2)}Ti_{0.14(2)}O₆

^eχ²= 1.42, R_{wp}= 5.82%, R_{exp}= 4.88%, R_B= 8.52%, Composition: Sr₂EuNb_{0.84(2)}Ti_{0.16(2)}O₆. Some Sr₃Ti₂O₇ is also present.

Table SI 2: Selected structural information for as prepared $\text{Sr}_2\text{EuNb}_{1-x}\text{Ti}_x\text{O}_6$ ($0 \leq x \leq 0.15$)

	$\text{Sr}_2\text{EuNbO}_6$	$\text{Sr}_2\text{EuNb}_{0.95}\text{Ti}_{0.05}\text{O}_6$	$\text{Sr}_2\text{EuNb}_{0.90}\text{Ti}_{0.10}\text{O}_6$	$\text{Sr}_2\text{EuNb}_{0.85}\text{Ti}_{0.15}\text{O}_6$
^a Tilt angle θ	16.5(6)	12.5(5)	13.8(6)	14.7(6)
^b Tilt angle φ	13.5(5)	12.2(5)	11.2(5)	11.5(6)
^c Tilt angle μ	12.2(5)	15.5(6)	15.1(5)	22.0(6)
Tolerance factor	0.9156	0.9160	0.9164	0.9168
^d B'-O(1) x 2	2.09(2)	2.08(2)	2.05(2)	2.13(2)
B'-O(2) x 2	1.96(2)	1.95(2)	1.97(2)	1.83(2)
B'-O(3) x 2	2.00(2)	2.07(2)	2.01(2)	1.93(2)
Average B'-O	2.02(1)	2.03(1)	2.01(1)	1.96(1)
^d B''-O(1) x 2	2.16(2)	2.23(2)	2.25(2)	2.33(2)
B''-O(2) x 2	2.32(2)	2.31(2)	2.26(2)	2.40(2)
B''-O(3) x 2	2.34(2)	2.19(2)	2.27(2)	2.36(2)
Average B''-O	2.27(1)	2.24(1)	2.26(1)	2.36(1)

^a B' = Nb/Ti, B'' = Eu

^b With [110]; $\theta = \frac{1}{2}$ [180-angle<B'-O(3)-B''>]

^c With [1-10]; $\varphi = \frac{1}{2}$ [180-angle<B'-O(2)-B''>]

^d With [001]; $\mu = \frac{1}{2}$ [180-angle<B'-O(1)-B''>]