Crystal structure and magnetism in $Nd_{1-x}Sr_xFeO_3$ ($0.1 \leqq x \leqq 0.9$)

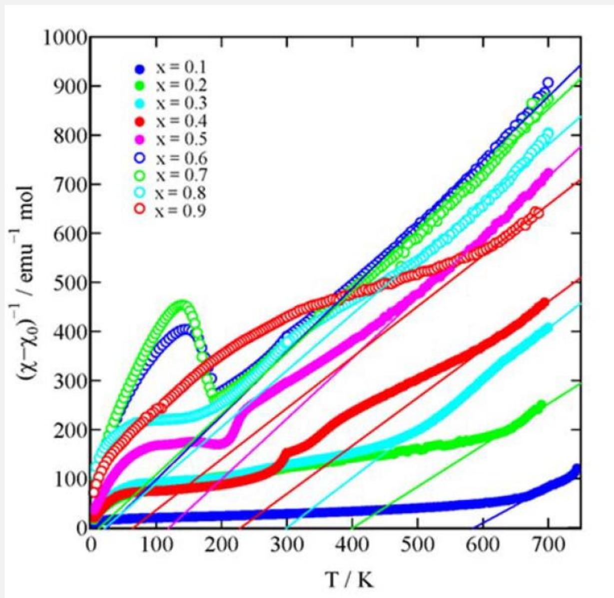
H.Nakatsugawa[1], Y.Kamatani[1], C.H.Hervoches[2]
[1]Yokohama National University, Japan, [2]Nuclear Physics Institute, Rež, Czech Republic

Introduction, the samples

Rare-earth orthoferrites $RFeO_3$ (R = La, Nd, Dy,…) are of particular interest due to their potential multiferroicity, magnetoelectric effects, and other functional properties.

In this study, several samples in the $Nd_{1-x}Sr_xFeO_3$ series ($0.1 \leq x \leq 0.9$) have been prepared, x-ray diffraction confirmed the obtention of single phase with a typical ABO_3 perovskite structure.

Magnetization measurements from 5K to 700K show weak antiferromagnetic behavior and paramagnetism following the typical Curie-Weiss law above 600K.



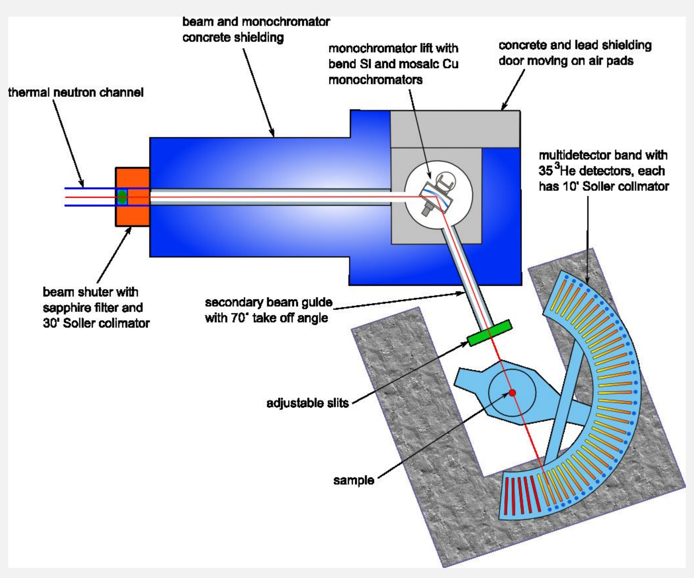
Inverse magnetic susceptibility vs T for $Nd_{1-x}Sr_xFeO_3$ ($0.1 \leq x \leq 0.9$)

In all samples, it is expected that Fe^{3+} is in low spin (LS) Fe^{3+} (t_{2g}^5); s = 0.5 or intermediate spin (IS) Fe^{3+} ($t_{2g}^4 e_g^1$); s = 1.5 and Fe^{4+} is in a low spin state (LS) Fe^{4+} (t_{2g}^4); s = 1.0.

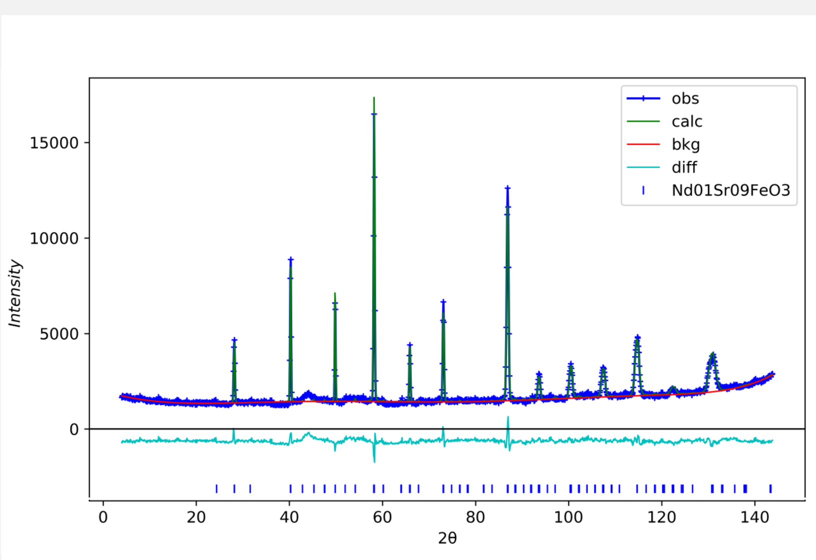
Thus, assuming that the mixed valence state of Fe^{3+} and Fe^{4+} is (LS Fe^{3+}_y IS Fe^{3+}_{1-y})$_{1-x}$ LS Fe^{4+}_x ($0.1 \leq x \leq 0.9$), the ratio of Fe^{4+} increases and that of Fe^{3+} decreases with increasing x.

In particular, the ratio of IS Fe^{3+}, which accounts for the largest proportion at x = 0.1 and 0.2, gradually decreases with increasing x. This decrease in the ratio of IS Fe^{3+} to the increase in x is expected to show a large correlation with the relaxation of the FeO_6 octahedron distortion.

The instrument: MEREDIT (MEdium REsolution neutron powder DIffracTometer)



Layout of the instrument



PND of $Nd_{0.1}Sr_{0.9}FeO_3$ (x = 0.9) at RT

Sample environment

- Variable temperature between 10 and 1300K with close cycle cryostat and vacuum or light furnace.
- Carousel for automatic sample exchange at RT.
- Euler goniometer for texture measurements.
- Deformation rig for mechanical testing.

Users open access portal: http://canam.ujf.cas.cz

Contact: hervoches@ujf.cas.cz

Neutron diffraction

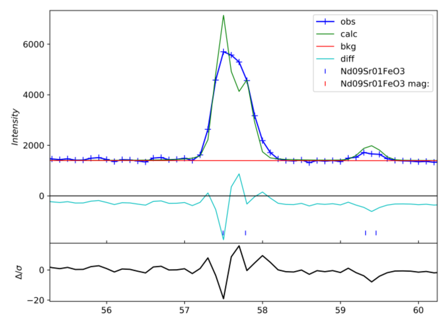
Powder neutron diffraction (λ=1.875 Å from a Si(311) monochromator) measurements were performed for all $Nd_{1-x}Sr_xFeO_3$ ($0.1 \leq x \leq 0.9$) samples at room temperature.

Results confirmed that the FeO_6 octahedron distortion is relaxed as x increases and approaches the crystal structure of the pseudo-cubic.

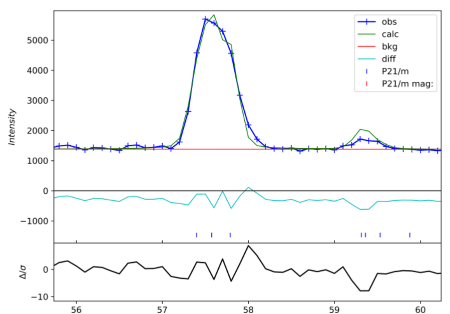
Neutron diffraction for all samples could be refined in orthorhombic space group Pnma but, for $0.1 \leqq x \leqq 0.2$, the quality is improved with space group $P2_1/m$, as seen with the fitting of 202 (and 20-2) reflections.

The following phase sequence is proposed:

- $0.1 \leqq x \leqq 0.2$ monoclinic $P2_1/m$
- $0.3 \leqq x \leqq 0.9$ orthorhombic Pnma

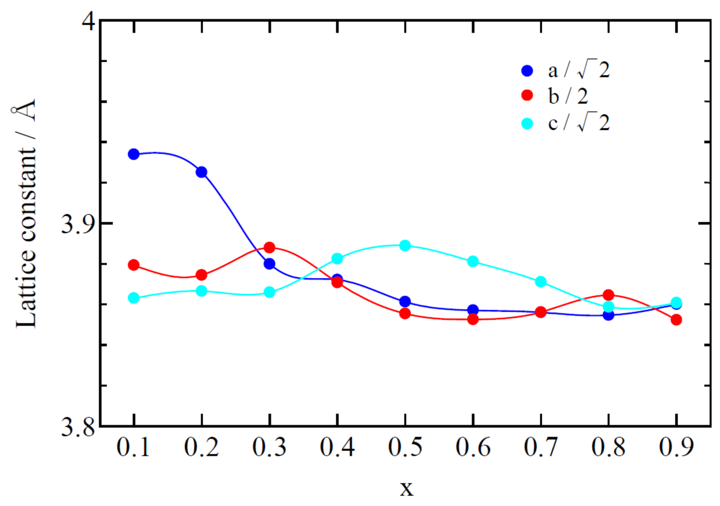


Pnma: 202 and 040

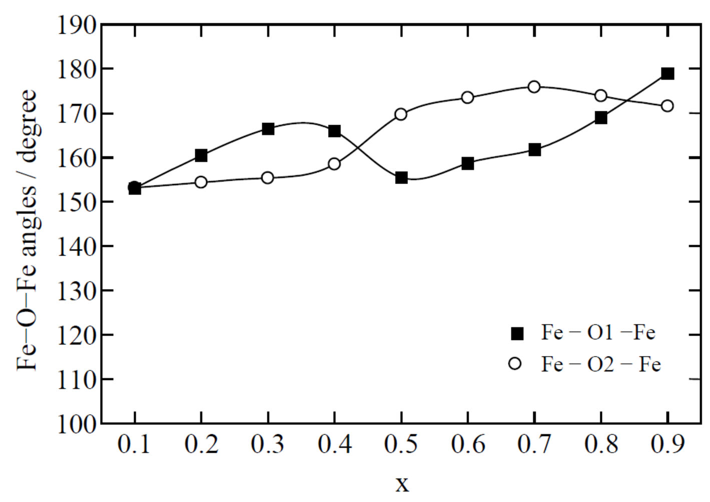


$P2_1/m$: 20-2, 202, 040

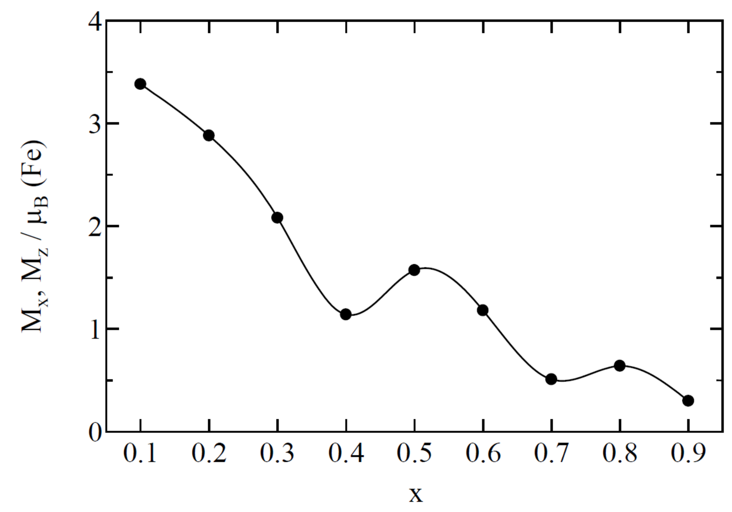
Crystal and magnetic structure change with x



Lattice parameters



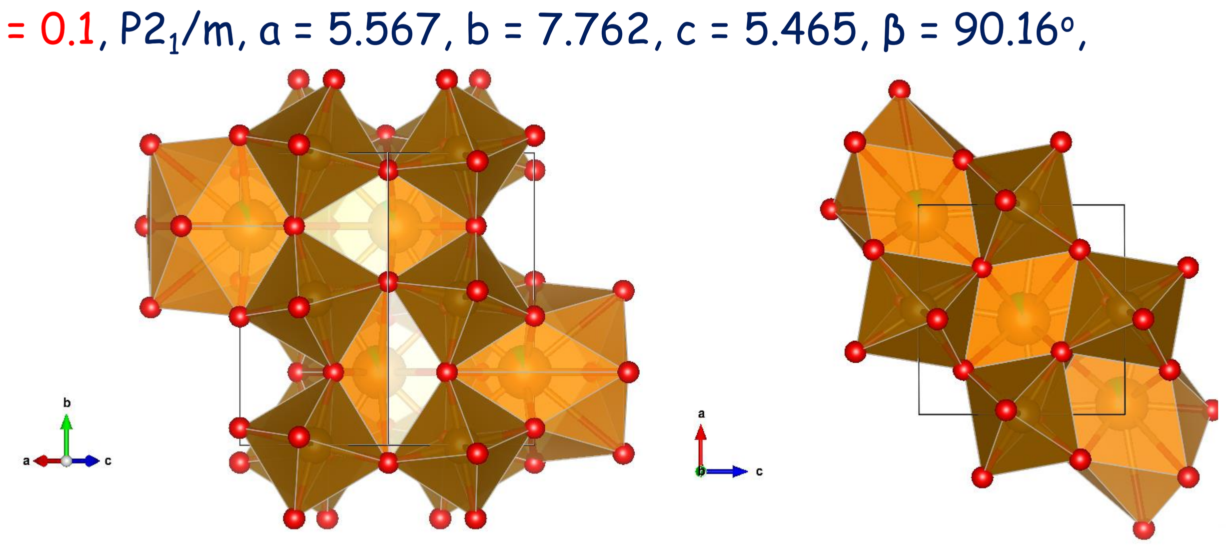
Fe-O-Fe angle

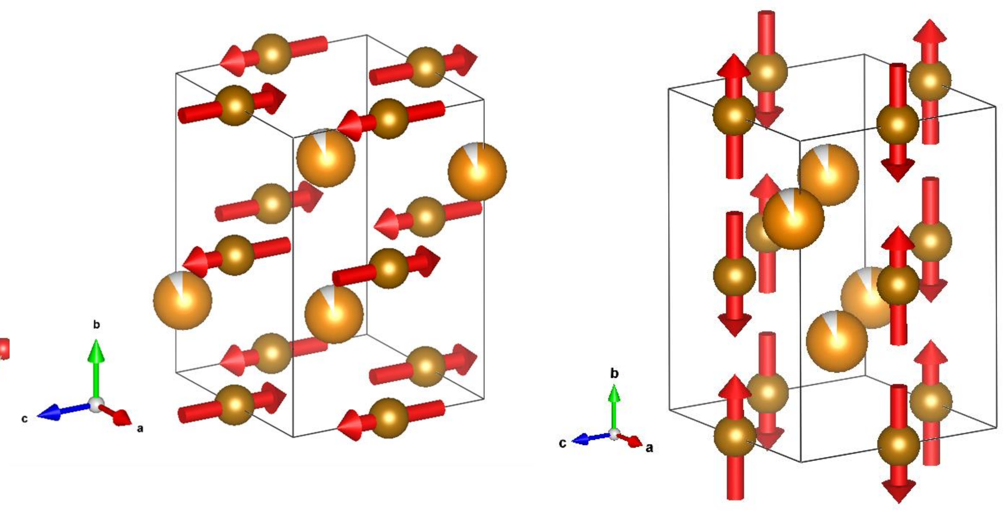


magnetization vs x

x = 0.1, $P2_1/m$, a = 5.567, b = 7.762, c = 5.465, β = 90.16°,

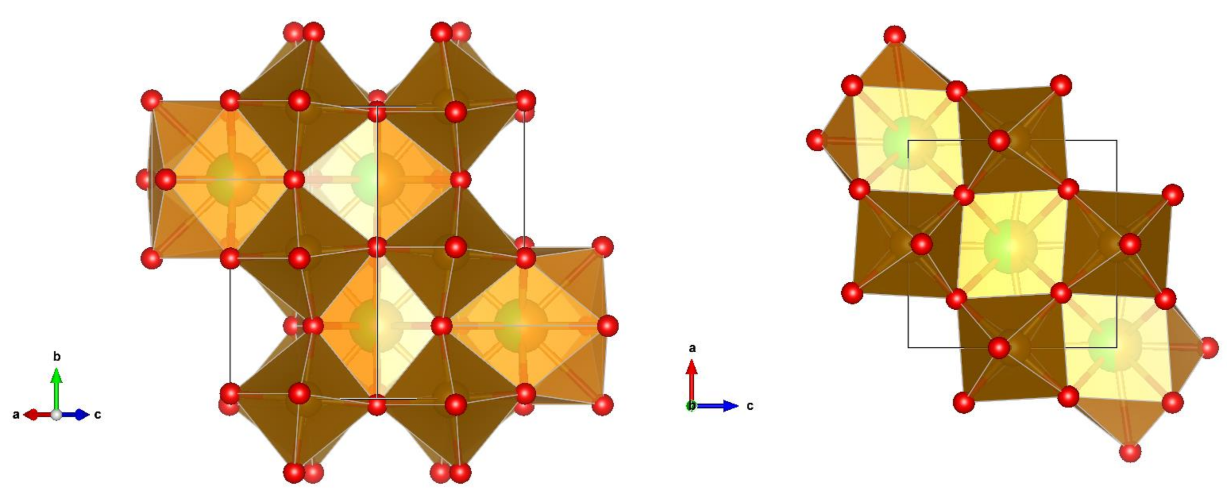
magnetic SG at RT: $P2_1/m$, Mz = 3.4, at 10K: $P2_1'/m'$, My = 3.7

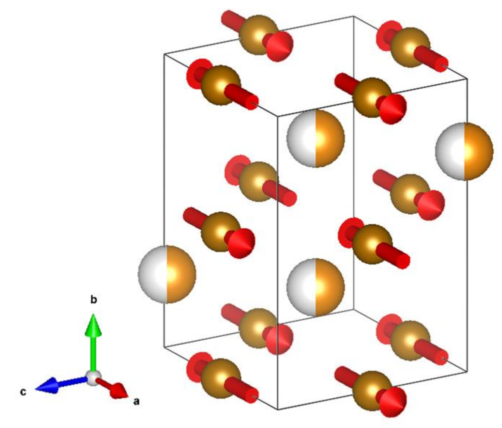




x = 0.5, Pnma, a = 5.456, b = 7.708, c = 5.489,

magnetic SG at RT: Pnma, Mx = 1.5





Drawing of the crystal structure showing tilting of the FeO_6 octahedron: in the (101) plane and about the b-axis,

Magnetic structure

Conclusions:

PND data of the $Nd_{1-x}Sr_xFeO_3$ samples were collected at RT for the whole series ($0.1 \leq x \leq 0.9$).

- The samples present antiferromagnetic ordering
- At room temperature, the magnetic spins order in the shorter axis, i.e.:
  - for $0.1 \leq x \leq 0.3$, the magnetic spins order in the c-axis direction (SG $P2_1/m$ or Pn'ma')
  - for $0.4 \leq x \leq 0.6$, the magnetic spins order in the a-axis direction (SG Pnma)
- At 10K, for $Nd_{0.9}Sr_{0.1}FeO_3$ (x = 0.1), the magnetic spins order in the b-axis direction (SG : $P2_1'/m'$)

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