ESRF	**Experiment title:** ***Phase Separation and Length Scales in CMR Manganites: $La_{5/8-x}Pr_xCa_{3/8}MnO_3$***	**Experiment number**: CH-1136
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Report

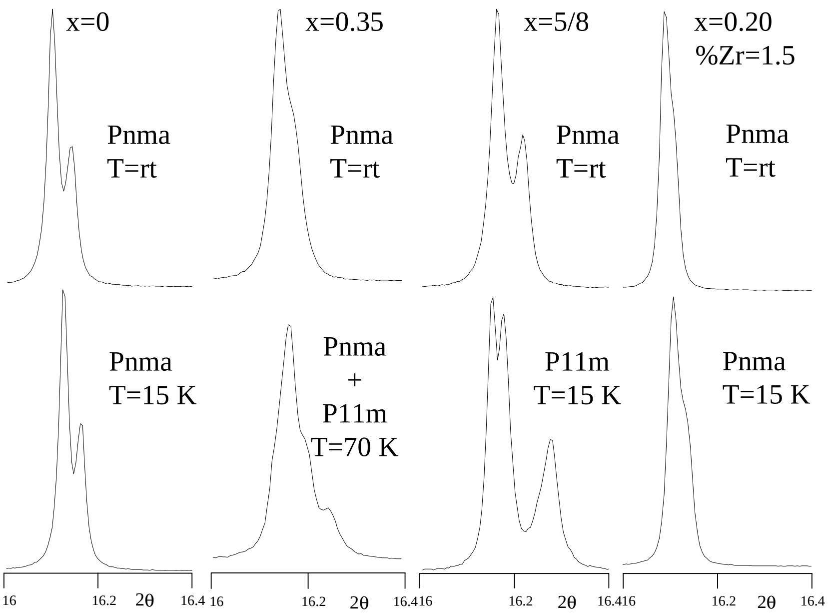
Phase separation/segregation is at the forefront of both theory and experiments in magnetic-conductors material science. $La_{5/8-x}Pr_xCa_{3/8}MnO_3$ series has been reported to display a very rich phase diagram at low temperatures. This archetype series displays metallic properties (x=0) and semiconducting behaviour (x=5/8) at low temperatures although the doping levels are the same. This shows the very important role of the chemical pressure or bandwidth tuning. Furthermore, for x=0.35 sample, a metal-insulator transition with Colossal Magneto-Resistance (CMR) and phase coexistence has been reported. At low temperatures, ferromagnetic-metal and antiferromagnetic-semiconducting-charge_ordered phases have been proposed to coexist. The doping level has been fixed to 3/8 because T_C is optimised at these compositions.

$La_{5/8-x}Pr_xCa_{3/8}MnO_3$ (x=0, 0.2, 0.35 and 5/8) samples were synthesised in Málaga. The magneto-transport characterization was done at ICMAB-CSIC (Barcelona). We have collected T-variable SXRPD data (@ BM16) to follow the phase(s) evolution, and T-variable NPD (@ D2B) to assign/determine the magnetic structure(s) of the phases. A short wavelength, 0.540092(1) Å, was used to minimise the absorption using capillaries of 0.5 mm of diameter ($\mu R \approx 1.4$ by assuming a packing 70 %). So, in this optminised conditions we collected 4 hours pattern (long scan) for full Rietveld analysis and 1 h patterns (short scan) to follow the unit cell and phase evolution with temperature. Patterns were collected over the temperature range 290-10 K. There was no problem with the beam and with the criostat, hence, the full planned experiment was carried out. We collected a NAC pattern at RT to have a reference for the microstructural study. Furthermore, There was enough time to collect patterns for an extra sample: $La_{5/8-x}Pr_xCa_{3/8}MnO_3$ (x=0.2) doped with 1.5 % of Zr in the B site of the perovskite.

$La_{5/8}Ca_{3/8}MnO_3$ does not undergo any structural phase transition on cooling and the M-I transition at 270 K is due to the ferromagnetic ordering of the manganese which induces the metallic-like behaviour. $Pr_{5/8}Ca_{3/8}MnO_3$ undergoes a phase transition on cooling at ≈ 220 K from orthorhombic P nma [a=5.44282(5)Å, b=7.65478(7)Å, c=5.42092(5)Å] to monoclinic P 11m [a=5.42537(5)Å, b=10.8807(1)Å, c=7.63041(8)Å and β=90.093(1)º at 100 K] with the a-axis of the P nma structure doubled. The splitting of the peaks is readily evident in the attached left figure. In the right figure is seen the superstructure peaks due to the charge-ordering. The coherence length is being evaluated by measuring the width of these peaks with temperature. The highest diffraction peak has 35000 counts, so the most intense CO peak has 0.4 %. This compound is semiconducting in the full temperature range. NPD data from D2B are fully compatible with the SXRPD data showing only ferromagnetic (**F**) peaks for $La_{5/8}Ca_{3/8}MnO_3$ and only anti-ferromagnetic (**AF**) peaks for $Pr_{5/8}Ca_{3/8}MnO_3$.

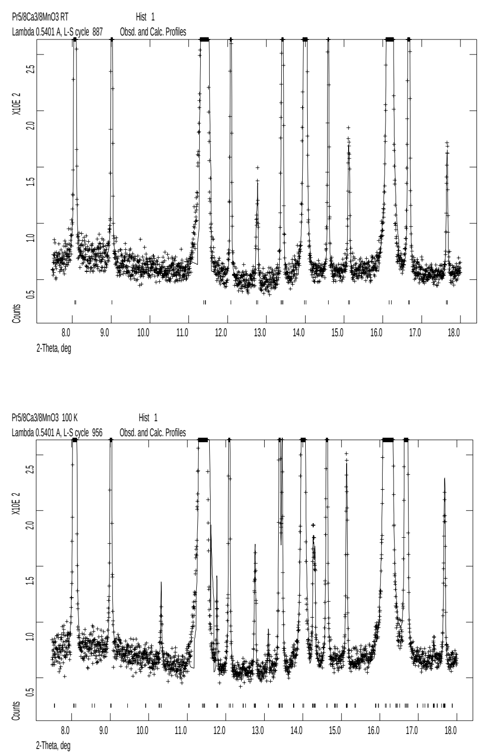
$La_{0.275}Pr_{0.35}Ca_{3/8}MnO_3$ undergoes a phase separation on cooling as seen in the left figure. D2B neutron data showed the coexistence of **F** and **AF** peaks although the phase separation "of the nuclear structure" was not seen due to the limited resolution of the neutron data. Here, we can assign the magnetic peaks to the corresponding nuclear structure. Furthermore, we are studying the evolution of the separated phases with temperature to understand the mechanism that drives this phase separation. There is a M-I transition at 110 K. We are evaluating whether the percolation threshold for the ferromagnetic phase is reached at this temperature or, as our data could demonstrate, a true phase transition occurs in the insulating fraction of the sample.

$La_{0.425}Pr_{0.20}Ca_{3/8}Mn_{0.985}Zr_{0.015}O_3$ undergoes a M-I transition at 80 K. SXRPD data showed no phase separation on cooling although D2B data showed the coexistence of **F** and **AF** peaks at 10 K. The magnetic peaks are being fitted with a canted magnetic structure.



Selected SXRPD data [(040) & (202) peaks] for $La_{5/8-x}Pr_xCa_{3/8}MnO_3$ (x=0, 0.35 and 5/8) and $La_{0.425}Pr_{0.20}Ca_{3/8}Mn_{0.985}Zr_{0.015}O_3$ on cooling.

x=0 ⇒ no change.
x=0.35 ⇒ phase separation.
x=5/8 ⇒ phase transition.
x=0.20 (1.5 % Zr) ⇒ no change.



Enlarged view of the SXRPD data for $Pr_{5/8}Ca_{3/8}MnO_3$ at RT and 100 K showing the ***fitted*** CO peaks

Full structural and microstructural data for this series will be reported elsewhere.