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Site specific magnetic frustration in oxygen vacancy ordered $La_xSr_{1-x}MnO_y$ manganites. A neutron powder diffraction study.

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Abstract – Oxygen vacancy ordered manganites with general formula $(La_xSr_{1-x})_mMn_mO_{3m-2}$ $0 \le x \le 0.2$ have been magnetically characterized using magnetization and neutron powder diffraction. Compounds with m=4 show the same magnetic ordering of previously known $Sr_4Mn_4O_{10}$ phase where application of Goodenough rules considering orbital ordering allows determination of magnetic ordering. For m=5 compounds orbital ordered Mn^{3+} cations in pyramids are coupled ferromagnetically in clusters of four pyramids that are antiferromagnetically coupled among them. Cations in octahedra separating pyramid clusters are coupled to two pairs of opposite moments. Depending on the charge of Mn in the octahedra this cation may or may not remain paramagnetic at 10 K as will be discussed.

Manganites of the $La_{1-x}Sr_xMnO_3$ series have been shown to display complex magnetic arrangements at low temperature, generally associated with charge and orbital ordered states [1]. When oxygen vacancies are added to the system the variety of structures increases with the addition of oxygen ordering phenomena [2]. At low oxygen contents $La_xSr_{1-x}MnO_{3-\delta}$ compounds with $x \le 0.2$ and $\delta \ge 0.4$ have been found to be formed by Mn^{3+} pyramids and $Mn^{3+/4+}$ octahedra with specific ordering patterns forming an homologous series [2]. Magnetic ordering in all-Mn^{3+}-pyramids $Sr_5Mn_5O_{10}$ ($SrMnO_{2.5}$) was reported by Caignaert [3] to be directed by simple Goodenough rules coupling when the complex orbital ordering pattern observed in this compound was taken into account. Using magnetization (Figure 1) and neutron powder diffraction we have performed the magnetic characterization of the $La_xSr_{1-x}MnO_{3-\delta}$ series to $(La_xSr_{1-x})_4Mn_4O_{10}$ and $(La_xSr_{1-x})_5Mn_5O_{13}$ compounds with $x \ge 0.8$ (equivalent to $La_xSr_{1-x}MnO_{3-\delta}$ with δ=0.5 and 0.4 respectively) showing oxygen vacancy, charge and orbital ordering and have verified that the same rules apply to the different magnetic ordering patterns analyzed. As a consequence of application of the Goodenough rules we have found that $(La_xSr_{1-x})_5Mn_5O_{13}$ (x=0.8 & 1) containing a 4/1 pyramid/octahedra ratio presents an antiferromagnetic structure where spins in pyramidal Mn^{3+} are ordered in ferromagnetic clusters of four pyramids coupled antiferromagnetically with every neighboring cluster leaving a disordered –frustrated- Mn octahedra at 10K as shown in Figure 2. Either Mn^{4+} or Mn^{3+} octahedra present in x=1 and x=0.8 compounds respectively have been found to be magnetically frustrated although different structural characteristics exist between both x=1 and x=0.8 compounds. This is, to the best of our knowledge, the first observation of geometrical magnetic frustration in a square lattice like the one present in these perovskite compounds.

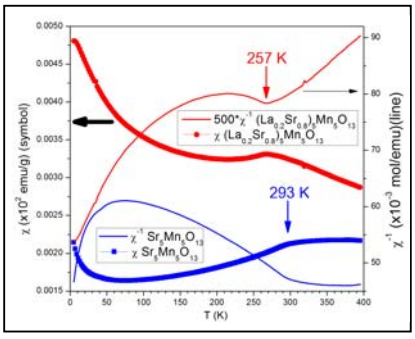

Figure 1: Magnetization of $(La_xSr_{1-x})_5Mn_5O_{13}$ x=0, 0.2.

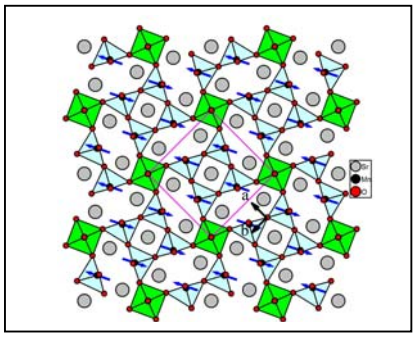

Figure 2: Magnetic structure of $Sr_5Mn_5O_{13}$.

References

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[3] Caignaert V., JMMM 166, 117 (1997).