ESRF	**Experiment title:** **Soft X-ray resonant scattering on the manganite $Nd_{0.5}Sr_{0.5}MnO_3$**	**Experiment number**: HE - 1874
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Names and affiliations of applicants (* indicates experimentalists):

J.Herrero-Martín[1*], J.García[1*], G.Subías[1*], J.Blasco[1], M.C.Sánchez[1] and S. Stanescu[2*]

[1]Instituto de Ciencias de Materiales de Aragón. CSIC-Universidad de Zaragoza, 50009 Zaragoza, Spain

[2]European Synchrotron Facility, B.P. 220, 38043 Grenoble, France

Report:

Near the half doping (i.e. x ≈ 0.5), where the formal valence on the Mn atom is +3.5, mixed valence manganites of the $RE_{1-x}A_xMnO_3$, $RE_{1-x}A_{1+x}MnO_4$ and $RE_{2-2x}A_{1+2x}Mn_2O_7$ (RE: rare-earth, A: alkaline-earth) families are the object of special attention because ionic charge (Mn^{3+}/ Mn^{4+}) and orbital ordering has been proposed as the fingerprint of the CE-antiferromagnetic insulating low-temperature phase. The classical model proposed to describe this low temperature phase is a one-dimensional zigzag chain of Mn atoms in the *ab* plane, coupled antiferromagnetically to each other. This model has been supported by the structural determination given by *Radaelli et al*. [1] showing that two non-equivalent crystallographic sites are present in the low temperature phase of $La_{0.5}Ca_{0.5}MnO_3$ for the Mn atom. In terms of the single ion picture, the tetragonal distorted octahedron is assumed to be $Mn^{3+}O_6$ while the $Mn^{4+}O_6$ one shows a nearly regular symmetry. Our recent x-ray resonant scattering (XRS) work [2] at the Mn K edge of a $Nd_{0.5}Sr_{0.5}MnO_3$ single-crystal agrees with the presence of these two types of non-equivalent Mn sites although a very small fractional charge segregation was deduced, according to $Mn^{+3.4}$-$Mn^{+3.6}$ intermediate-valence states.

In this experiment, we have measured the (0½0) and (½00) reflections (Fig.1) at the Mn $L_{2,3}$ absorption edges of the same $Nd_{0.5}Sr_{0.5}MnO_3$ single-crystal via XRS in the soft x-rays. The single crystal, grown at Zaragoza University using a floating zone furnace, was cut and polished in the $[001]_{cubic}$ plane. It turned out to be twinned in the [100/010] directions but both (½00) and (0½0) reflections could be measured separately as the correspondent diffraction peaks are angularly resolved. Thus, this becomes the first work in which orbital and spin contributions are decoupled at the same temperature.

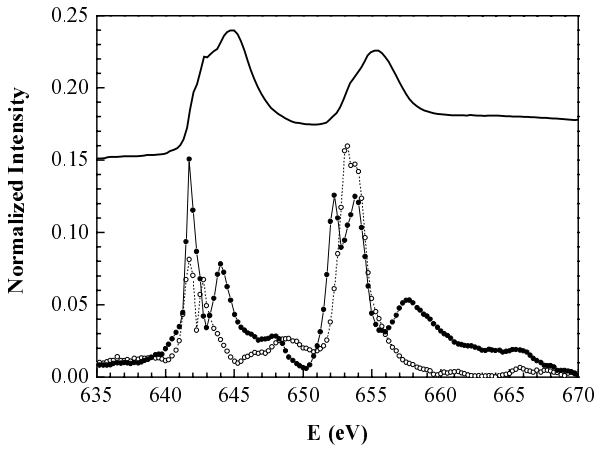

Figure 1. Energy dependence of the two reflections studied for σ incident radiation: (0½0) is denoted by filled circles and (½00) is marked by open circles. The absorption spectrum (solid line) is shown for comparison at the top, displaced in the vertical axis.

Measurements were taken at different azimuthal angles and temperatures ranging from 30 K to 180 K, crossing the so-called charge-ordering transition at $T_{CO} \approx 150$ K. Both (0½0) and (½00) reflections were present at T=30 K. The intensity of the two reflections decreases with increasing temperature, being completely disappeared at 170 $\pm$ 10 K. We observed four main peaks in the (½00) and (0½0) spectra, which are coupled in pairs at energies close to the Mn L_3 and L_2 edges, respectively. These peaks did not however correlate between the two reflections, either in shape or in energy position, except for the first peak, at 641.7 eV. This suggests a different origin for both reflections. Our experimental results are consistent with those reported in recent publications [3,4]. The azimuthal evolution (ϕ: azimuthal angle) was identical for the two studied reflections, following a sinusoidal with maxima at ϕ=90º and null minima at ϕ=0º. The same azimuthal behavior was observed for the (05/20) reflection in our previous XRS study at the Mn K edge [2].

The comparison with our previous XRS experiment at the K edge in the same compound suggests that the observed resonances at the so-called orbital ordering (0½0) reflection should be due to the anisotropy of the d-projected density of states coming from the tetragonal distortion of the anisotropic MnO_6 octahedron and it cannot be described only using a simplistic ionic model. Up to now, the small number of XRS experiments carried out in half-doped manganites at the Mn $L_{2,3}$ edges have been always interpreted within this ionic context. In such a way, in our system and following our notation, half-integer reflections of the form (0 *k*/2 0), *k* odd, have been described as originating from the differences in the filling and orientation of the 3d atomic orbitals at the Mn sites. Meanwhile, (*k*/2 0 0) reflections, *k* odd, have been proposed to have a magnetic origin due to the net electron spin orientation on these sites.

Summing up, anisotropic and magnetic resonant reflections have been observed in the half doped $Nd_{0.5}Sr_{0.5}MnO_3$ in the energy range of soft x-rays. However, a more detailed analysis, including a comparison with theoretical simulations, would be useful to fully confirm the origin of the observed resonances.

[1]: P. G. Radaelli *et al*., *Phys. Rev. B* **55**, 3015 (1997)

[2]: J. Herrero-Martín *et al*., *Phys. Rev. B* **70**, 024408 (2004)

[3]: K. J. Thomas *et al*., *Phys. Rev. Lett*. **92**, 237204 (2004)

[4]: S. S. Dhesi *et al*., *Phys. Rev. Lett*. **92**, 056403 (2004); S. B. Wilkins *et al*., *Phys. Rev. B* **71**, 245102 (2005); U. Staub *et al*., *Phys. Rev. B* **71**, 214421(2005)