



	Experiment title: Polarization analysis of charge ordering (030) and (300) reflections in Nd_{0.5}Sr_{0.5}MnO₃ by X-ray resonant scattering at the Mn K-edge	Experiment number: HE - 1388
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Report:

We have investigated the low temperature phase (well below $T_{CO} \approx 150K$) of a $Nd_{0.5}Sr_{0.5}MnO_3$ single crystal by x-ray resonant scattering (XRS) at the Mn K-edge of the (300), (030) and (05/20) reflections. We have proposed a semi-empirical structural model [1], which nicely explains the occurrence of resonant scattering at the so-called charge (CO) and orbital (OO) ordering reflections.

Our single crystal was cut and polished to a flat (100)_{cubic} surface. This surface of the crystal resulted to be twinned, both (100) and (010) domains being detected. At $T=60$ K, strong resonances were observed for the $\sigma-\sigma'$ channel at the (300) and (030) reflections and for the $\sigma-\pi'$ channel at the (05/20) reflection (Fig.1). Neither a $\sigma-\pi'$ contribution in the former reflections nor a $\sigma-\pi'$ contribution in the latter were detected. As a function of the photon energy, resonances appeared near the Mn K edge. A non-resonant intensity was observed at energies below the absorption edge in the case of the (300) and (030) reflections whereas it was zero for the (05/20) reflection. This background is the Thomson contribution, a structural modulation coming from a small motion of the atoms out of the $Ibmm$ symmetry. The intensity of these reflections was also studied as a function of the azimuthal angle (rotation around the [001] direction). The same azimuthal behavior of the scattered intensity at resonance is observed for all the studied reflections, following a sinusoidal with π period.

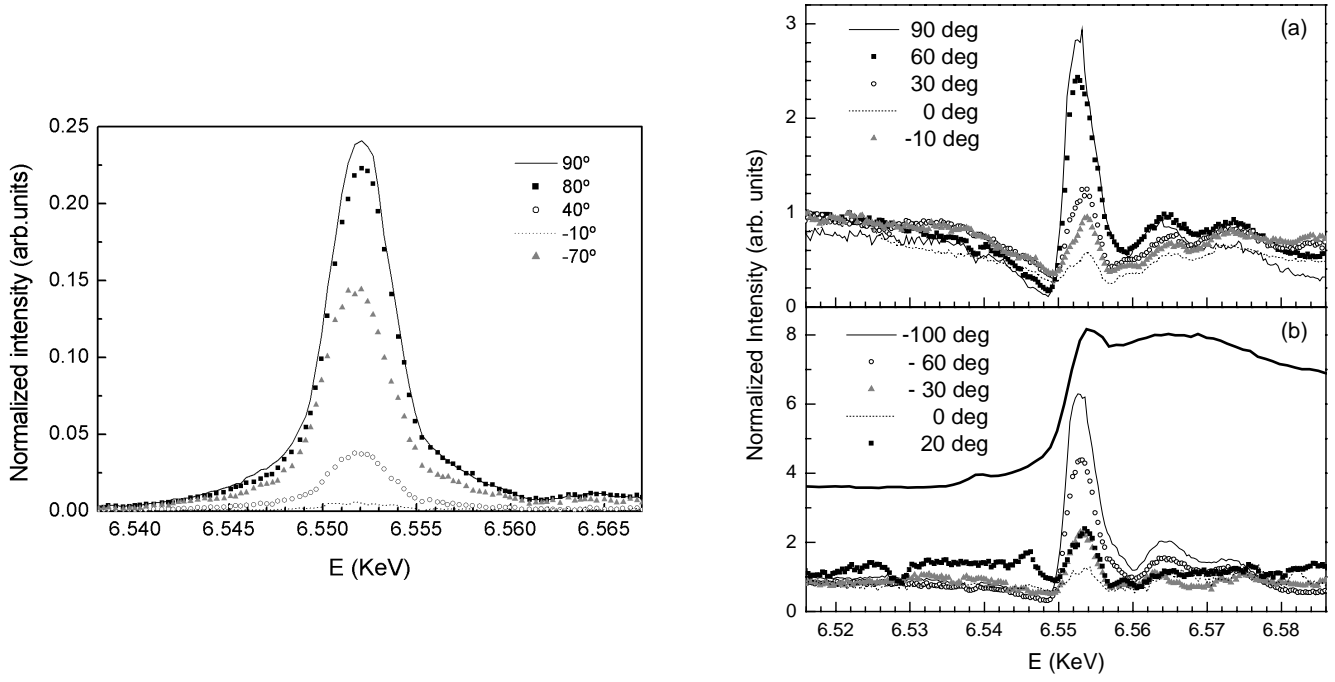


Figure 1. Intensity of the (05/20) σ - π' [left] and (300) and (030) [right] reflections as a function of the energy at different azimuthal angles. On the right, panel (a) shows data for the (300) reflection and panel (b) for the (030) one in the σ - σ' channel. The reference for the azimuthal angle ($\phi, \phi'=0$) corresponds to the crystallographic directions [010] for (300) and [100] for (030) and (05/20) reflections, respectively. The fluorescence spectrum is also shown for comparison (thick solid line).

The detailed analysis using a tensorial formalism, based on the anisotropy of the atomic scattering factors contributing to the structure factors of the studied reflections, nicely reproduced the experimental azimuthal and polarization dependence. By means of a semiempirical approximation, we were successful in fitting the experimental XRS spectra introducing three parameters: chemical (δ_{chem}) and anisotropic (δ_{anis}) energy shifts and Thomson scattering terms $C_{h(k)}$. We have used the experimental non-polarized XANES spectrum to obtain the different atomic scattering factors for the tensor components. The best-fit values of these parameters point towards a much smaller charge disproportionation between the two species of Mn atoms in the cell than the ionic approximation of 1 electron. Thus, it is not appropriated to speak in terms of Mn^{3+}/Mn^{4+} ions but rather preferably $Mn^{3.4+}/Mn^{3.6+}$. On the other hand, anisotropy related to the tetragonal distortion of the so-called $Mn^{3+}O_6$ octahedra plays a very important role and it is the only responsible of the appearance of half-integer reflections such as (05/20) in the σ - π' channel. Nevertheless, resonances at the (300) and (030) reflections also have a contribution from this anisotropy (δ_{anis}) as well as from the charge disproportionation (δ_{chem}).

The so-called COO phase transition has been therefore explained as a structural phase transition where two non-equivalent Mn sites, tetragonal distorted and nearly regular octahedron, are present in the low-temperature phase and a partial segregation of charge takes place as a consequence. This is, however, far from the ionic approximation Mn^{3+}/Mn^{4+} .