XMaS the UK-CRG	**Experiment title:** **Azimuthal Scans of charge and orbital ordering in CMR manganites using the polarisation analyser**	**Experiment number:** 28-01-071
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Report:

The perovskite manganites have been of great interest recently due to their extraordinary physical properties, especially colossal magnetoresistance (CMR).[1] The charge, spin and orbital degrees of freedom each play an important role in the electrical and magnetic properties of the transition-metal oxides[2], but it is only recently with that it has been possible to observe orbital order with 3rd generation synchrotron sources.[3]

We have performed resonant x-ray scattering studies and azimuthal scans at the Mn *K*-edge on a high quality single crystal of $Nd_{0.5}Sr_{0.5}MnO_3$ grown at Bell Laboratories. Studies carried out on the magnetic and transport properties have shown that the system undergoes a transition from a paramagnetic to a ferromagnetic state at 240 K followed by a 1st order transition at 160 K to a charge ordered anti-ferromagnetic insulating state[4]. At the Mn *K*-edge a resonance was observed by us in the charge and orbital ordering reflections due to a sharp increase in the anomalous scattering factors and the sensitivity of the 4p orbitals to the e_g electron orientation. In addition at the absorption edge both the charge and orbital ordering have tensor qualities and this results in a $\sin^2\Psi$ angular dependence in the intensity.

The crystal was mounted with the [1,1,0] axis surface normal on the 11-axis Huber diffractometer and the sample rocking curve measured on a Bragg peak was approximately 0.1 °. The incident x-ray energy was tuned to the Mn *K*-edge and the X-ray beam was plane polarised in the σ plane. Using a copper (220) polarisation analyser crystal it was possible to observe the scattered signal in both the σ and π channels, making it possible to separate any spin or orbital signals from the charge background. Using a closed cycle Displex cryostat the sample was cooled through the charge ordering transition to a temperature of 100 K. We located charge order (CO) peaks at a modulation of (0.5, 0, 0) and (0, 0.5, 0) in the σ channel, and carried out energy scans and azimuthal scans on the (1.5, 2, 0) CO peak over a range of 210° around the scattering vector.

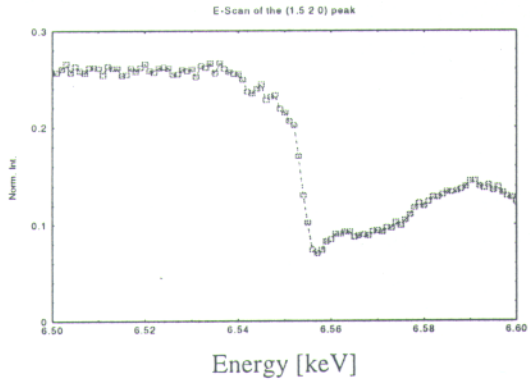

Figure 1 Energy Scan of the (1.5, 2, 0) Distortion

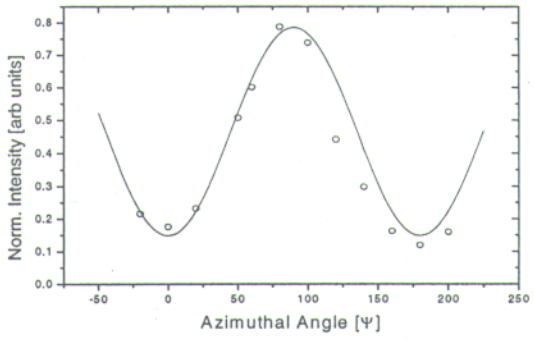

Figure 2 Azimuthal scan of the (1.5, 2, 0)

These satellites were initially believed to be charge order caused by the increase in the anomalous scattering factors at the Mn *K* absorption edge but were in fact due to a lattice distortion associated with the Jahn-Teller ordering of the oxygen octahedra. An energy scan carried out in the range 6.5 keV to 6.6 keV showed no resonance as shown in Figure 1 and it was observed in the σ channel, and it exhibited the same behaviour as an energy scan carried out on the (2, 2, 0) Bragg peak, with a drop of intensity at the absorption edge. This behaviour shows that the peaks with wavevector (0.5, 0, 0) are not due to the changes in the anomalous scattering factors of the Mn^{3+} and Mn^{4+} ions around the Mn *K*-edge. Instead they are caused by the tilting and stretching of the oxygen octahedra around the Mn^{3+} ion associated with the Jahn-Teller distortion. The intensity of the distortion shows a clear $\sin^2\Psi$ angular dependence at the resonant energy of 6.553 keV, but unlike charge order the scattered intensity exhibits the $\sin^2\Psi$ angular dependence off-resonance at 6.52 keV as well and at both energies the intensity at the maxima was approximately 4 times greater than that at the minima of the azimuthal scan.

The azimuthal dependence can be explained by considering the shape of the oxygen octahedra, the 2 Mn-O bonds along the c direction are elongated compared to the other 4 bonds. This means the electron density of the distortion is not symmetrical and when rotating around the scattering vector with [1,1,0] surface normal an azimuthal dependence is seen in the intensity as the orientation of the octahedra changes with respect to the incoming beam. We did not observe orbital ordering most likely because its predicted position was at the same modulation as the distortion and it would be obscured by the 3% leak through from the distortion in the π channel. We were unable to observe the charge ordering and why this is the case is unclear but it could be it is not possible to observe it along the [1,1,0] axis whereas elsewhere when it was observed the [1,0,0] axis was surface normal.

We have demonstrated the presence of a distortion at (0.5 0 0) modulation that shows no energy resonance at the Mn-*K* edge but does exhibit a $\sin^2\Psi$ azimuthal dependence associated with the elongation of the Mn-O bonds along the c-direction but we were unable to see the charge or orbital order observed elsewhere. Clearly more investigation is required into the interplay between the charge, orbital and Jahn-Teller order to allow the properties of the manganites to be better understood.

References

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