ESRF	**Experiment title:** X-ray resonant magnetic scattering during a spin-reorientation transition	**Experiment number**: HE1537
Beamline:	**Date of experiment**: from: 27/01/04 to: 3/02/04	**Date of report**: 21/9/04
Shifts:	**Local contact(s)**: P. Bencok, S. Stanescu	*Received at ESRF:*

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Report:

This report concerns the use of beamtime allocated for experiment number HE1537. Unfortunately, the proposed experiments could not be carried out because the stray fileds from the magnet were enough to destroy any stripe domains that may have been created. Instead, we report here alternative backup experiments that were performed.

Perovskite manganites, such as $La_{0.5}Sr_{1.5}MnO_4$, have enabled pioneering studies of the delicate balance between spin, charge and orbital degrees of freedom [1-3]. At room temperature the Mn sites have an average valence of +3.5, but below ~217 K charge ordering (CO) has been proposed leading to two inequivalent sites identified as Mn^{3+} and Mn^{4+} [1-3]. Hund's rule coupling implies that the Mn^{3+} $3d^4$ site has three electrons in the $t_{2g\uparrow}$ level and one electron in the twofold degenerate $e_{g\uparrow}$ level. The degeneracy in the $e_{g\uparrow}$ can be lifted by collective Jahn-Teller distortions (JTD) of the oxygen octahedra or by antiferromagnetic spin ordering [4] leading to an anisotropic 3d charge distribution at the Mn^{3+} sites known as orbital ordering (OO). On the other hand, even the existence of CO is disputed while OO has also been proposed as a consequence of spin ordered zigzag chains within a band-structure model. Unfortunately, the controversy surrounding OO remains difficult to resolve largely because existing probes of OO are too indirect.

X-ray diffraction is mainly sensitive to the rather isotropic electron distribution, but tuning the photon energy to an absorption edge gives enhanced sensitivity to valence states. Resonant x-ray diffraction, involving virtual excitations from core to valence states, can then probe the anisotropic valence charge density allowing forbidden diffraction peaks to appear [1]. Recently, Mn K-edge resonant diffraction from $La_{0.5}Sr_{1.5}MnO_4$ implied that OO is established at the same temperature as CO (217 K) and that it is a precursor to the complex antiferromagnetic spin ordering observed at lower temperatures [3]. However, it is still not clear whether OO arises from spin ordering, from the JTD, or from a combination of the two. Soft x-ray resonant diffraction (SXRD) is developing into a versatile probe of structural and magnetic phenomena and was used to study OO in $La_{0.5}Sr_{1.5}MnO_4$.

Measurements were done using a five-circle diffractometer, operating at a base pressure of 1×10^{-10} mbar, and equipped with a Si diode detector mounted behind variable rectangular apertures. The (110) surface was polished with diamond paste to a flat surface and mounted in the UHV diffractometer without further *in situ* surface preparation. The intensity of the (¼ ,¼, 0) diffraction peak across the Mn $L_{2,3}$ edge and at 134 K is shown in Figure 1. At the Mn $L_{2,3}$ edges the (¼ ,¼ ,0) forbidden reflection has significant intensity due to the resonance from OO. The inset shows a (¼+Δq, ¼+Δq, 0) scan of the forbidden reflection at a photon energy of 643.8 eV and at T=134 K (open circles). Figure 1 demonstrates the presence of a detailed structure with two regions of high intensity below and above ~650 eV which are due to virtual excitations

from the $2p_{3/2}$ and $2p_{1/2}$ core levels to unoccupied 3d states. Each region has five main contributions with a broad feature at ~664 eV.

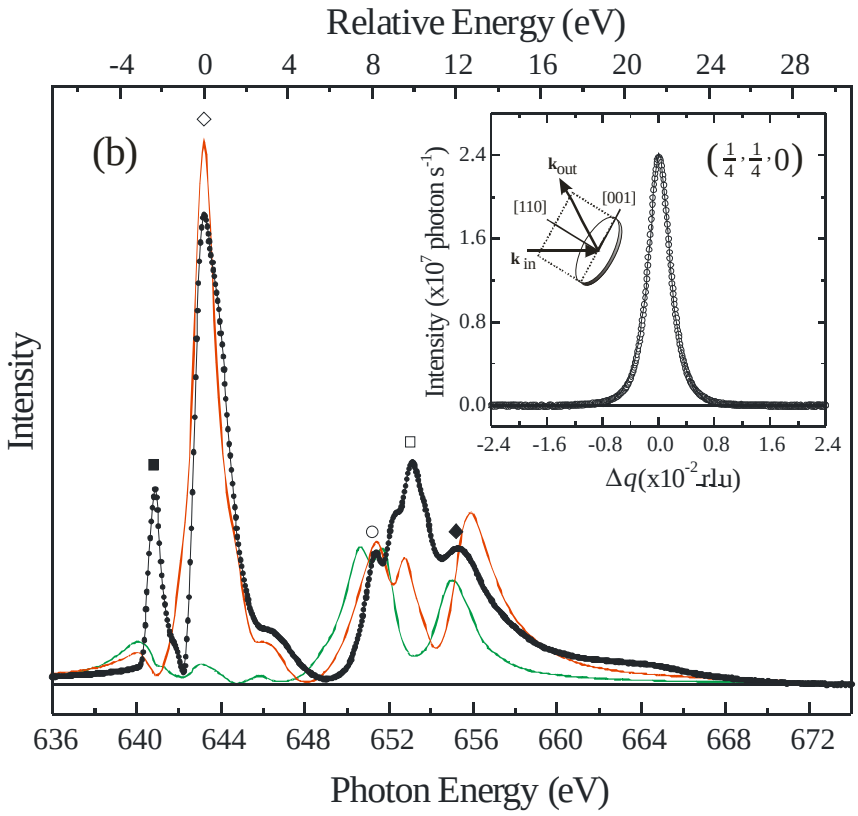

Figure 1 The energy dependence (solid circles with line) of the (¼ ,¼, 0) peak recorded over the Mn $L_{2,3}$ edges at 134 K and the calculated spectrum for large (red line) and small (green line) JTD. The inset shows a (¼+Δ*q*, ¼+Δ*q*, 0) scan of the forbidden reflection arising from OO at a photon energy of 643.8 eV and at T=134 K (open circles). The solid line is a Lorentzian fit tothe peak. The experimental geometry is also shown.

To determine the origin and the type of OO present, the energy dependence of the diffraction peak was compared to ligand-field atomic-multiplet calculations. Figure 1 shows the energy dependence of the OO peak (red line) calculated using the parameters representing a significant JTD. These parameters correspond to an almost complete OO of the $3d_{z^2\text{-}r^2}$. As seen, the agreement between the experimental and calculated spectra is good. In order to isolate the effects of JTD on the OO, calculations were also performed for parameters which reduce the effects of the JTD. The calculated energy dependence is shown by the green line in Fig.1. The removal of the JTD results in an almost complete drop in intensity for the main peak at the L_3 edge and the shoulder at ~3eV higher in energy. The contributions at the L_3 edge are therefore mainly influenced by JTD whereas at the L_2 edge contributions arising from spin correlation effects are more relevant [5,6]. This, therefore, represents the first direct evidence of OO at the Mn 3d states arising from spin correlations.

In conclusion, soft x-ray resonant diffraction has allowed new insights into the process of OO in $La_{0.5}Sr_{1.5}MnO_4$ [6]. A comparison of the energy dependence of an intense diffraction peak, arising from 3d OO, with theoretical calculations implies that JTD together with spin correlations drive the OO process. It is expected that the complex relationship between the charge, orbital and spin degrees of freedom can now be explored in greater detail using the increased sensitivity of SXRD.

References

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