ESRF	**Experiment title:** Electric and magnetic response of Mn^{2+} in the multiglass $(Sr,Mn)TiO_3$ probed with hard x-ray linear and circular dichroism	**Experiment number**: HE-3146
Beamline: ID 12	**Date of experiment**: from: 24.11.2009 to: 01.12.2009	**Date of report**: 01.03.2010
Shifts: 18	**Local contact(s)**: A. Rogalev, F. Wilhelm	*Received at ESRF:*

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Report:

The aim of this experiment was to study the magnetic and electric response of a $(Sr,Mn)TiO_3$ (SMnT) polycrystal under electric and magnetic field to unravel the off-central position of the Mn ion in the multiglassy phase using x-ray linear dichroism (XLD). It was chosen to prepare a thin Au electrode on either side of the SMnT polycrystal to apply an external electric field to switch on and off the electrical polarization of the SMnT which should be linked to an off-centrality of the Mn ion and therefore a characteristic XLD. This XLD should have been studied as a function of external electric and magnetic field.

During the beamtime the initial experimental approach was to use the external electrical field as lock-in signal instead of chopping the synchrotron light. Figure 1 shows exemplarily the XANES and related XLD signal recorded at the Ti K edge. The XLD was in this case derived by the difference between the XANES

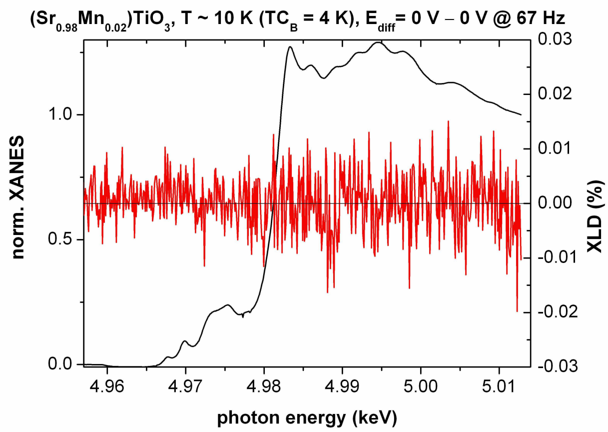

Figure 1: XANES and XLD spectra at the Ti K-edge of SMnT at 10 K, recorded at E=0V (10 spectra averaged)

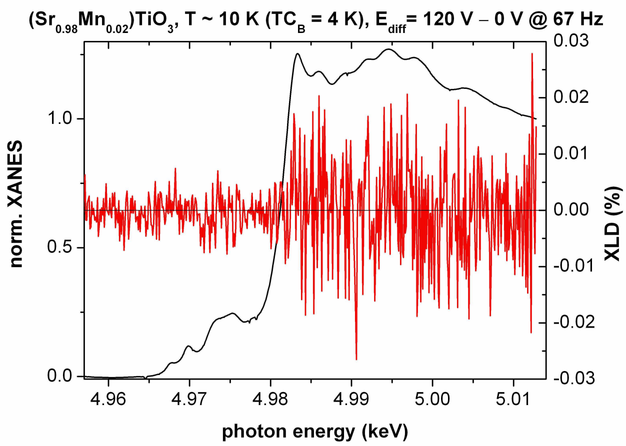

Figure 2 XANES and XLD spectra at the Ti K-edge of SMnT at 10 K, recorded at E=120V and 67 Hz (10 spectra averaged)

with 0V and 0V switched at a frequency of 67 Hz, i.e. the test signal if the beamline components were stable. 10 spectra were recorded at about 10 K and subsequently averaged. A clear zero-line with an excellent noise level of 0.01% of the edge jump is visible in Figure 1. It should be noted that switching the electric fields at 2 Hz leads to an energy shift visible in the XLD spectra which were a factor of 2-3 larger than the zero line in Figure 1. The same experiment was repeated and the difference was now taken between an external electric field of 120 V and 0 V, i.e. this signal should reflect an electric field-induced off-centrality of the Ti. Surprisingly, the only effect of the electrical field is, that the noise level underneath the main absorption is increased by about a factor of 3 without any specific spectral shape. The source of this is currently unclear but it may indicate that the glassy state may lead to the situation that each time the field is switched on and off, the system goes into a slightly different microscopic state, which also differs in the respective XLD signal.

Although the used electric fields should fully saturate the electric response beyond the hysteretic region, this possibility cannot be fully ruled out. It should be noted, that these studies at the Ti K edge were only performed because unexpected problems occurred while recording the XLD at the Mn K-edge, which will be discussed in the following.
Figure 3 shows the XANES and XLD spectra recorded at the Mn K-edge at 10 K as a difference between 180 V and 0 V with a modulation frequency of 67 Hz. For this single XLD spectrum 68 (!) spectra were averaged to yield this excellent signal to noise ratio of 0.1%. However, no XLD signature is visible. Both XANES and XLD are indicative of a very low Mn concentration well below the expected nominal concentration of 2%. Having recorded the (nonexistent) XLD shown in Figure 3, the experiments shown in Figures 1 and 2 were conducted as test-experiments for the reliability of the findings at the Mn K-edge. The time was used to cool down the Si drift detector (SDD) to increase the sensitivity of the detection system. Having found out, that there may be a reproducibility-issue related to modulating the sample with an electrical field of 67 Hz, it was decided to record the XLD spectra under static electrical fields with the usual chopper for lock-in detection. This detection mode also makes the detection with the slow SDD feasible. The sample had to be shifted to the 6 T magnet for this purpose and the sample alignment turned out to be extremely difficult using the SDD. From the absolute count rate of the Mn $K_{\alpha 1,2}$ radiation it could be concluded that within the probing depth of the x-rays, the Mn concentration should be about 0.02%, i.e. two orders of magnitude lower than expected. This can explain the difficulties in recording XLD spectra at the Mn K-edge with good signal-to-noise-ratio. Unfortunately, by then it was already Monday, i.e. only one night was left. Thus, it was decided to probe the XLD using the SDD only under a static electric field of 180 V with and without a magnetic field of 6 T. Since the magnetic field control stalled during the last night, only one XLD spectrum with 180 V and 0 T could be recorded which derived from an average of three individual spectra and it is shown in Figure 4. A clear XLD of 2% at the Mn K-edge is visible which can be taken as a first proof-of-principle experiment for the chosen experimental approach. The large size of the XLD suggests that it should be also visible using the detection mode in Figure 3. However, as the findings at the Ti K-edge suggest, there may be reproducibility-issues when cycling SMnT at 67 Hz. The origin of these issues is unclear so far an in contrast to integral measurements. On the other hand, the findings in Figure 4 clearly demonstrate the existence of an off-centrality of the Mn ion. Due to lack of time we could not check, whether the XLD stems from the electric field or not. However, having resolved the unexpected experimental issues, we have at least established a proof of principle and the XLD spectrum can serve as reference for a future, more systematic study varying both electric and magnetic field to record different XLD spectra.

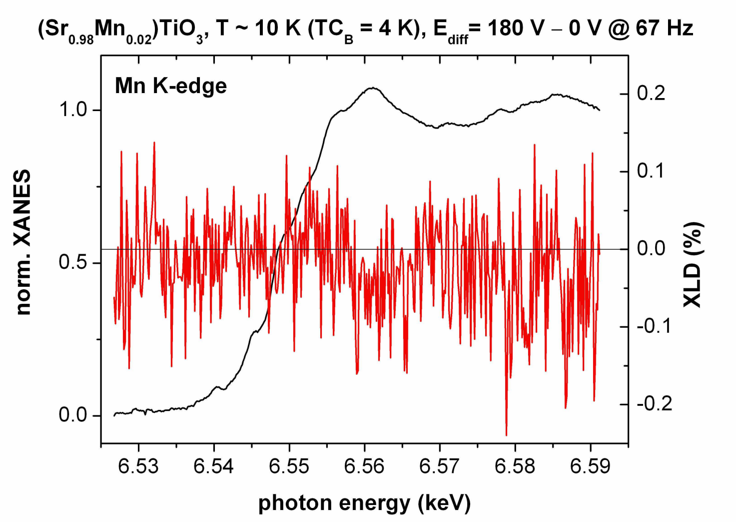

Figure 3 XANES and XLD spectra at the Mn K-edge of SMnT at 10 K, recorded at E=180V at 67 Hz. Note, that the huge number of 68 (!) spectra were averaged to derive the shown (nonexistent) XLD.

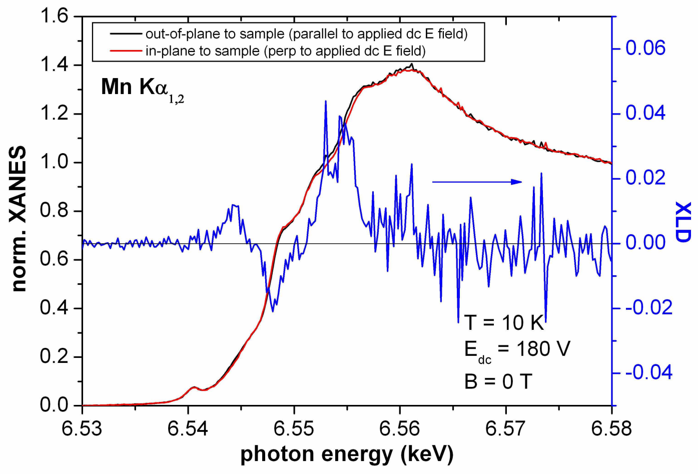

Figure 4: XANES and XLD spectra at the Mn K-edge of SMnT at 10 K, recorded at static E=180V using the Si drift detector. (3 spectra averaged). No external magnetic field was applied.