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| 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-3438 |
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## Report:

The aim of this experiment was to study the local structural and magnetic properties of Mn-doped $SrTO_3$ (STO) in external electric and magnetic fields using x-ray linear dichroism (XLD) and x-ray magnetic circular dichroism (XMCD) being a continuation of HE-3146. The available set of samples was extended to Mn doping levels of 2%, 3% and 5%. All samples were bulk polycrystals and were covered with a 50nm thick Au electrode forming a plate-capacitor geometry. The samples were all studied at low temperatures of ~8K, i.e. well-below the glass-temperature of these combined spin- and dipole-glasses. As pointed out in the previous report of HE-3146 we had to use the Si-drift detector to be able to measure the XLD of the 2% Mn:STO sample due to the large background signal of the high Ti content of the samples. During the beamtime, however the drift-detector was not operational due to electronic problems thus restricting the experiments to the 5% and 3% Mn doped STO samples. Because of the same reason, it was only possible to study the XLD signal but not the XMCD signal which is much smaller. Two different types of XLD experiments were carried out during this experiment: (i) using linear polarized light and switching the external E-field on/off and (ii) using the quarter-wave plate to flip the linear polarization for a given constant E-field. (i) yielded a smaller, more noisy XLD if any (see also Report HE-3146); therefore we restricted ourselves to (ii). Since for the 5% Mn-doped STO sample the XANES indicates that most of the Mn was not incorporated on Sr but on Ti lattice sites, most attention was given to the 3% Mn-doped STO sample and these results shall be presented in the following. Note, that each XLD spectrum which is shown in the followinig already represents an average of 6 indvidual XLD scans across the Mn K-edge with 4 flips of the polarization at each energy point, thus comprising a very long averaging time to yield a resonable signal-to-noise ratio of the XLD.

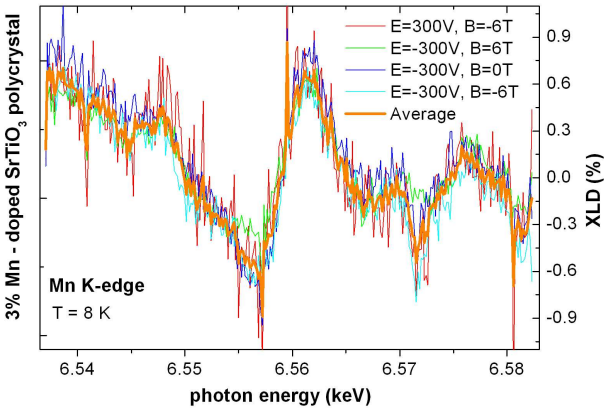


*Figure 1: XLD spectra at the Mn K-edge of 3% Mn:$SrTiO_3$ recorded at 8 K in applied E field of +/-300 V for all B-fields of +/-6T and 0T and the resulting average.*

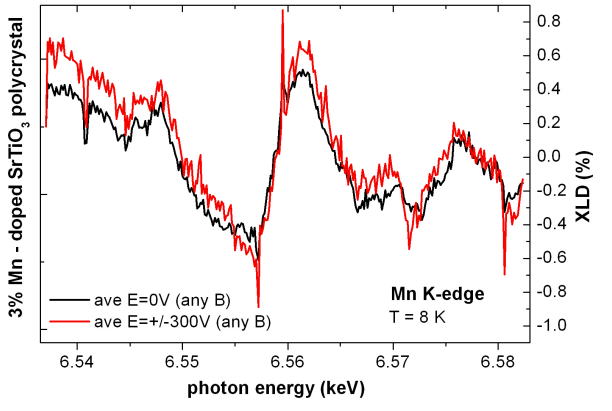


*Figure 2: Average of XLD spectra recorded at E=0V and E=300V (from Fig. 1) over all externally applie B-field values.*

Figure 1 shows exemplarily various XLD spectra at the Mn K-edge recorded at low temperatures (8K) in an applied electric field of -300 V. For three XLD spectra a magnetic field of +6T, -6T or 0T was applied in addition, respectively. Since the magnetoelectric coupling in this multiglass is expected to be of the order of $B^2E^2$, for a B-field of -6T a reversed E field of +300V was measured as well yielding no significant discrepancy, thus corroborating the quadratic nature of the E-field dependence. To extract the influence of the applied electric field on the local structural distortion around the Mn ion, all four resulting XLD spectra were averaged (thick orange line in Fig. 1) irrespective of the applied magnetic field.
The same experiment was performed in zero electric field in addition at the same magnetic fields or +6T, -6T and 0T, resepctively (not shown). The average of the latter is shown together with the one from Fig. 1 in Figure 2. At first glance only small discrepancies are visible suggesting, that the electrical field has only a weak influence on the local distortion of the Mn-doped STO. However, the presence of an XLD signature itself already indicates local symmetry breaking around the Mn ion even without any applied field which may be explained by either self-poling of the material while cool-down or intrinsic stress of the crystallites. The actual origin of this "spontaneous" XLD is not yet fully understood.
Furthermore, a comparable experiment was carried out to extract the influence of the magnetic field, i.e. measuring the XLD either in +/-6T or in 0T and averaging over the electrical field of -300V and 0V, respectively. These XLD spectra were averaged accordingly and the respective outcome is shown in Figure 3. These two XLD reflect the influence of the external magnetic field on the local structural distortion of the Mn. As in Fig. 2 also in Fig. 3 the discrepancy between the two XLD spectra is small. Note, that the sign of either field should not play any role because the magnetoelectric coupling scales like $B^2E^2$, i.e. only the changes with respect to zero electric or magnetic field can be studied.
Figure 4 displays the differences of the XLD spectra without and with field of Fig. 2, i.e. electric field, and Fig. 3, i.e. magnetic field, respectively. A significant change is observable in both cases around 6.555 keV, i.e. around the maximum of the XANES spectrum of the Mn K-edge (see Report HE-3146). In both cases the XLD spectra change in a very similar manner upon applying either an electric or a magnetic field.
This photon energy was then used to try to record XLD(E) curves either in applied magnetic field or without magnetic field (not shown). These curves were intended to serve as corroboration of the $E^2$-dependence of the magnetoelectric properties. Unfortunately, these were done at the very end of the beamtime leaving only little time for sufficient statistics, so that the resulting XLD(E) curves were rather noisy. Nonetheless, we could demonstrate the principal feasibility to record the electrical field dependence of the local distortion, i.e. a physical quantity proportional to the (electric) polarization, by means of an element selective spectroscopic technique. This demonstrates the viability of the chosen approach and could be refined using single crystals.
In summary, indications for a magnetic-electric coupling in Mn-doped STO could be found using a local spectroscopic probe. The local environment of the Mn ion was shown to be altered by applying either an electric or a magnetic field at 8 K using XLD at the Mn K-edge. The spectroscopic signatures reflecting the altered local structural properties of the Mn ion are similar for both electric and magnetic fields. Interestingly, the applied fields are rather large, so that the glass-like properties of this material should not be visible any more but only the magneto-electric coupling.

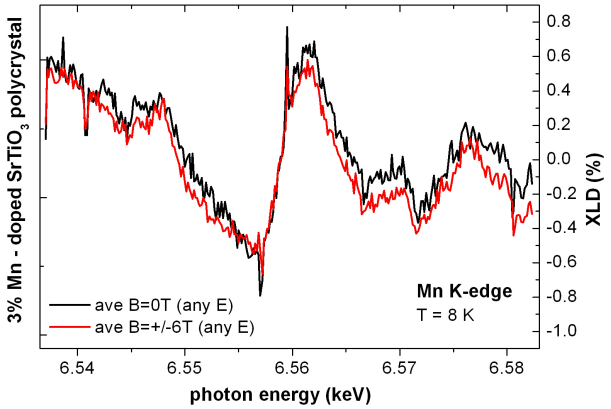


*Figure 3: Respective XLD spectra as in Fig. 2 but averaged over all E-field values for B=0T and B=6T.*

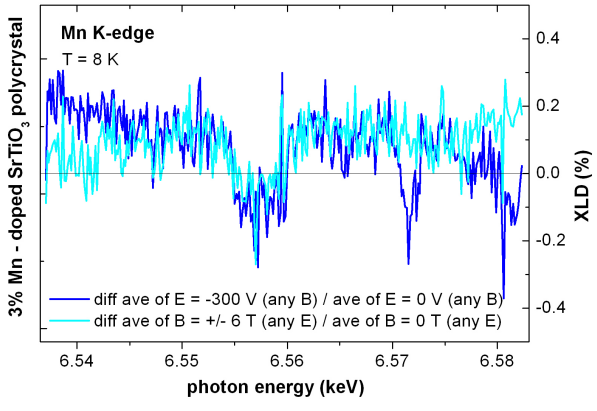


*Figure 4: Difference of the averaged XLD spectra of Fig. 2 and Fig. 3, respectively, showing the change in the XLD spectra caused by the electric and the magnetic field. The resulting spectral feature is similar thus revealing magneti-electric coupling in Mn:STO.*