ESRF	**Experiment title:** Probing the Mn incorporation and magnetism in magnetically doped topological insulator Bi_2Se_3 using XLD and XMCD	**Experiment number**: HC-1225
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Report:

The aim of the proposal is to study incorporation of Mn into the topological insulator (TI) Bi_2Se_3 by x-ray linear dichroism (XLD) and to determine the associated magnetic properties using x-ray magnetic circular dichroism (XMCD) up to high magnetic fields. During the beamtime two sets of Mn-doped Bi_2Se_3 samples grown on BaF_2(111) substrates were investigated, one with 200 nm thickness an one with 500 nm both having various Mn concentrations. For all samples the XANES and XLD were recorded at the Bi $M_{5/4}$-edges (Figure 1), for the Se K-edge (Figure 2) and the Mn K-edge (Figure 3). The XLD for the two former edges was derived from measurements using circular light and taking the difference between grazing an normal incidence. Especially at the Bi M-edges this leads to a strong background in the resulting XLD spectra which is mainly due to the anisotropic fluorescence background of the Ba of the substrate and it is visible in Fig. 1 that this background is less pronounced for the thicker samples. On top of this background a clear oscillatory behavior in the XLD is visible, which can be also seen at the Se and more clearly at the Mn K-edge. Preliminary simulations of the spectra using the FDMNES code can reproduce this thus being indicative of this oscillation being linked to the structure of this TI with its stacked quintuple layers of Bi-Se separated by the Se van-der-Waals gap. Especially at the Se K-edge the spectra were full of parasitic Bragg peaks of the substrate, so that only for the thick Bi_2Se_3 films the spectra could be somehow corrected for those unwanted artifacts still leading to a bumpy appearance of the XLD, see Figure 2. The comparison of the FDMNES simulations to these spectra is thus not meaningful and complicates determination of the incorporation of the Mn into the Bi_2Se_3. Figure 3 shown the XANES and XLD recorded at the Mn K-edge which could be recorded using the quarter-wave plate to flip the linear polarization thus being devoid of strong background.

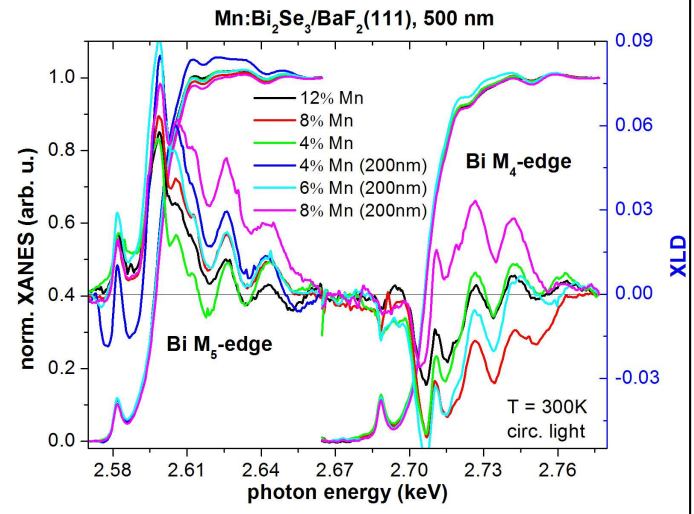

Figure 1: XANES and XLD at the Bi $M_{5/4}$-edges.

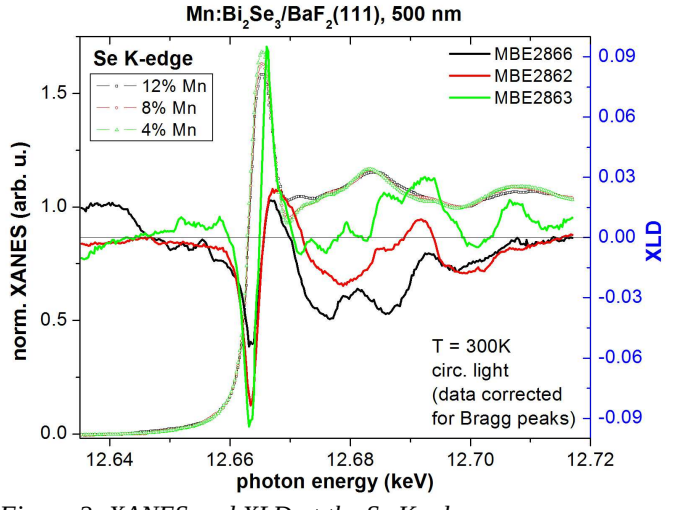

Figure 2: XANES and XLD at the Se K-edge.

All XLD spectra in Figure 3 clearly exhibit the oscillatory behavior at higher photon energies and a more or less pronounced sharp feature at the edge. Preliminary simulations reveal that this may be indicative of a substitutional incorporation of the Mn on Bi sites in the quintuple layer of Bi_2Se_3. However, the overall agreement with the simulations is not very satisfactory (thus not shown) and there are indications of at least some Mn being located within the Se van-der-Waals gap. Only if clean spectra are available for all edges, these indications may be consolidated and substantiated. We want to note, that we have also taken some preliminary data on comparable Mn-doped Bi_2Te_3 films (not shown) which point toward a different incorporation of Mn in the selenide and telluride, respectively. The data are however not complete and also suffer from the strong background of both the Ba fluorescence as well as Bragg peaks from the substrate. Finally, XCMD spectra have been taken at the Mn and also Se K-edge. The Mn K-edge XMCD is shown in Figure 4 together with a field dependence of the XMCD up to 17 T. The latter is compared to the scaled SQUID data which were routinely recorded for all samples. First, spectral shape of the XMCD closely resembles the XLD in Figure 3, however, the XMCD reverses clearly with magnetic field and the peaks in the XMCD are also shifted compared to the XLD in energy (not shown). Since the Bi_2Se_3 samples show an in-plane easy axis an angular dependent XMCD study would be very interesting to correlate the anisotropic electronic states in the XLD with the anisotropic magnetic states in the XMCD. However, recording the XMCD in normal incidence was not possible since the films were too thin (500 nm) to sufficiently suppress the background; under grazing incidence the XMCD spectra look quite fine. The Se K-edge XMCD is not shown because we were running out of time towards the end of the beamtime so that we could not record the field dependence and thus an important information is missing, if we picked up a reliable magnetic signal, since the two XMCD spectra recorded with opposite magnetic field direction as a difference of the two helicities show a strong superposition of a magnetic as well as nonmagnetic (i.e. residual XLD like) contribution. Therefor the field dependence would be crucial to provide clear evidence for a magnetic polarization of the Se and thus a good evidence for incorporation of Mn next to Se in the Bi_2Se_3 samples. Turning back to the XMCD data in Fig. 4 there are two important observations possible: (i) towards the end of the XMCD spectrum there is a clear peak in the XCMD which is not caused by a Bragg peak. We were also able to measure the field dependence of this feature and it exactly follows the behavior of the XMCD peaks right at the edge. In fact this field dependence is also contained in the averaged data shown in the inset. We attribute this feature to a double electron excitation (simultaneous excitation of the K and M edge) which may be very interesting by itself but does not contribute to the actual objective of the proposal. (ii) The XMCD(H) curve in the inset of Fig. 4 shows that the magnetization does not saturate up to 17 T which can be taken as a first evidence for the presence of antiferromagnetic interactions in these ferromagnetic samples. These may be due to a secondary Mn containing antiferromagnetic phase, e.g., Mn-Se like configurations stemming from some Mn in the Se van-der-Waals gap. However, since these time-consuming measurements were only done for one Mn concentration so that definite answers cannot be given yet.

In summary, although we were able to record many XANES, XLD and XMCD spectra for a large set of samples, most of the data suffer from background issues. These shall be overcome by thicker films on BaF_2 or using other substrates. We have first indications for a mostly substitutional incorporation of Mn on Bi sites as well as antiferromagnetic interactions underlining the interesting magnetic properties of the TI Bi_2Se_3.

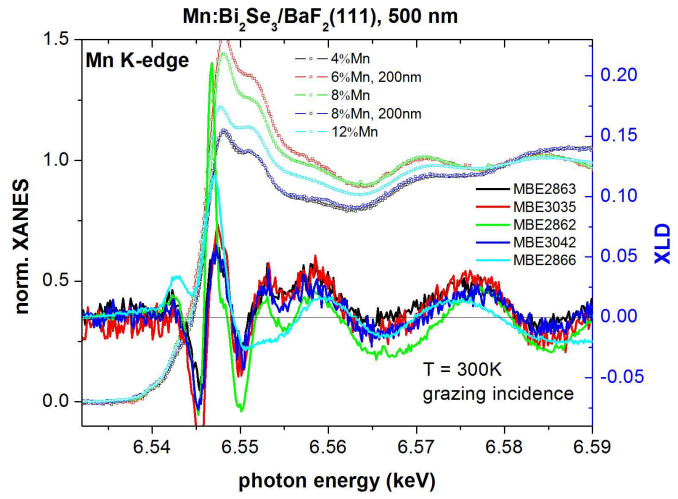

Figure 3: XANES and XLD at the Mn K-edge.

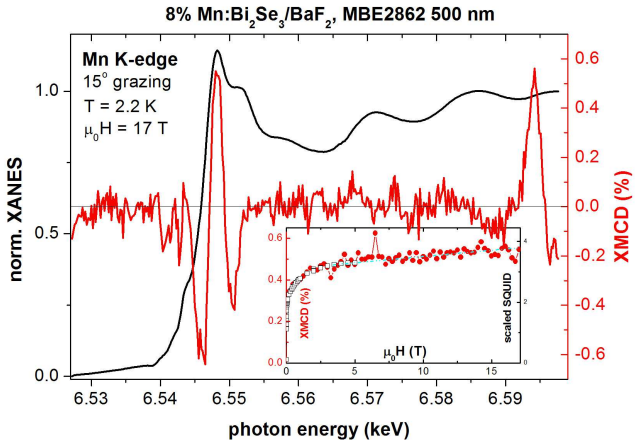

Figure 4: XANES and XMCD at the Mn K-edge. The inset shows the field dependence of the XMCD together with the scaled SQUID data.