ESRF	**Experiment title:** Probing the magnetic polarization of the As-sublattice in the alpha- and beta-phase of MnAs	**Experiment number**: HE-2861
Beamline: ID 12	**Date of experiment**: from: 18.02.2009 to: 24.02.2009	**Date of report**: 12.08.2009
Shifts: 18	**Local contact(s)**: A. Rogalev, F. Wilhelm	*Received at ESRF:*

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Report:

The aim of this experiment was to clarify the magnetic structure of β-MnAs by studying an eventual magnetic polarization of the As site in the β-phase, i.e. at elevated temperatures. Other than in previous attempts, MnAs powder sealed in epox should have been used to avoid excessive background signals from the otherwise used GaAs substrates. Precharacterization using SQUID has revealed satisfying magnetic properties of the sealed MnAs powder. However, during the beamtime it turned out, that the absorption edges found for Mn and As K-edges were all characteristic of oxidized MnAs material. Tenn different sample types using different epoxy, preparation temperatures and powders have been tested, but none exhibited sufficiently clean metallic Mn (Fig. 1) and As spectra (Fig.2) which could be used to study XMCD at the As K-edge in a meaningful manner, since oxidation may uncontrollably alter the magnetic state of MnAs. These measurements took about 25% to 33% of the beamtime

It was therefore decided to study "backup samples" instead. For that a number of Co-doped ZnO films grown by reactive magnetron sputtering (RMS) and pulsed laser deposition (PLD) were available. The RMS samples were of different Co concentration such as 10% and 15%, whereas the PLD samples were with 5% Co. For all types of samples a pair of paramagnetic (PM) and superparamagnetic (SPM) samples were available which was induced by altered preparation conditions. X-ray linear dichroism was recorded for a few of them, to check for the local structural quality. In could be confirmed for two pairs of samples that PM is correlated with maximum possible (compared to previous beamtimes HE-2553, HE-2714) XLD signal whereas SPM samples show a reduced XLD. In addition for one set of samples extended x-ray absorption spectra (EXAFS) have been recorded, which other than XLD do not exhibit a clear difference between PM and SPM samples.

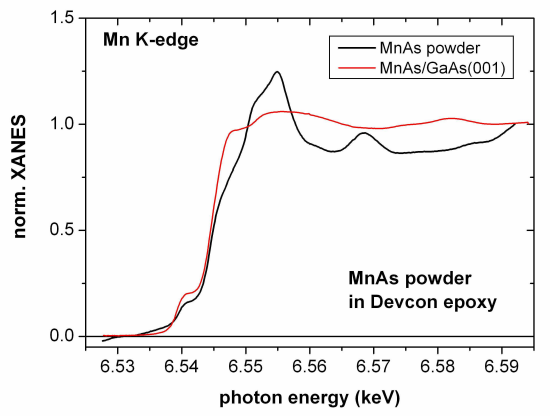

Figure 1: XANES spectra at the Mn K-edge of MnAs powder in epoxy compared to an epitaxial film of MnAs on GaAs (001) revealing the powder to be oxidized

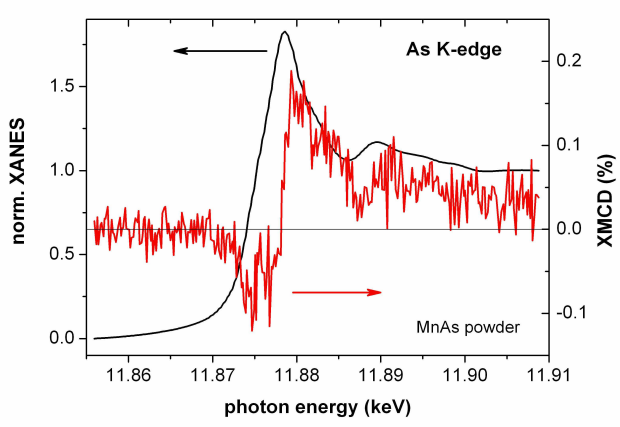

Figure 2: XANES and respective XMCD spectra recorded on MnAs powder in epoxy at 300 K. A small XMCD signal is visible but the XANES reveals that the As is oxidized as well.

Most of the remaining beamtime was used to magnetically characterize pairs of PM SPM samples using x-ray magnetic circular dichroism and element specific M(H) curves at the Co K-edge. Figure 3 exemplarily shows XANES, XMCD and element specific M(H) curves of a PM 5% Co-doped ZnO sample grown by PLD, which shows the full XLD signal (not shown). A clear dichroic signal is visible at the pre-edge feature which is characteristic of Co^{2+} in tetrahedral coordination. The field dependence reveals pure PM behavior. In addition, the field dependence of the XMCD signal in the valley between pre-edge feature and the onset of the main absorption is probed. At this photon energy metallic Co has its most prominent dichroic signal. The absence of any field dependence of the XMCD at this energy demonstrates the absence of metallic-like magnetic contributions from the Co dopant. In contrast, Fig. 4 shows exemplarily a SPM 15% Co-doped ZnO sample grown by RMS under reduced oxygen partial pressure. This sample shows reduced XLD at the Co K-edge (not shown). The XMCD at the pre-edge feature is reduced and it shows PM-like field dependence. However, probing the field dependence of the XMCD at the photon energy of the valley, a small but systematic dichroic signal is recorded which shows a characteristic step-like behavior around the origin. This signature is indicative of a small metallic-like contribution to the overall magnetic properties, which provides microscopic evidence for the source of the SPM behavior of such types of samples. Note, that also the valley in the XANES itself is less pronounced in Fig. 4 compared to Fig. 3, which also indicated increasing metallicity of the Co dopant atoms. This fraction of the beamtime together with parts of HE-2860 has therefore been used contribute to a comprehensive study which summarizes characteristic spectral features of hard-x-ray-based techniques such as XANES, XLD and XMCD for the system Co-doped ZnO. We could compare PM-SPM pairs of samples stemming from four different institutions to establish meaningful quality indicators based on XANES, XLD and XMCD to discriminate between the intrinsic properties of Co-doped ZnO which is PM and signatures for extrinsic contributions from phase separation which leads to SPM. The results as exemplarily shown in Figs. 3 and 4. were used to probe the presence or absence o metallic Co contributions to the magnetism by the respective XMCD signal at the characteristic photon energy. The energy and site specificity of synchrotron-based techniques can unravel small FM-like contributions to an overall PM signal, which is virtually impossible by integral magnetometry. In addition, microscopic information is provided about the local structural and electronic properties. Our comprehensive manuscript is currently under review at Nature Materials and the above-discussed measurements were included in that work.

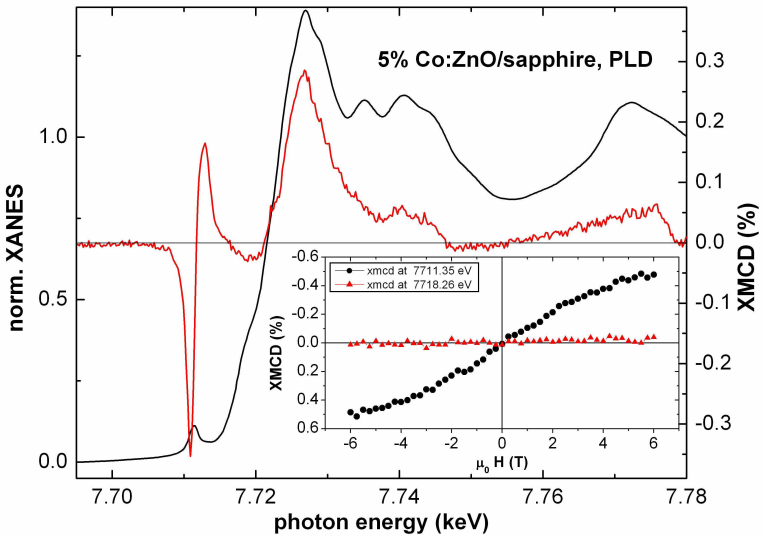

Figure 3: XANES and respective XMCD spectra of a PM 5% Co:ZnO sample grown by PLD. The M(H) curves in the inset reveal PM at the pre-edge feature and no signal in the valley, where the XMCD of metallic Co can be probed.

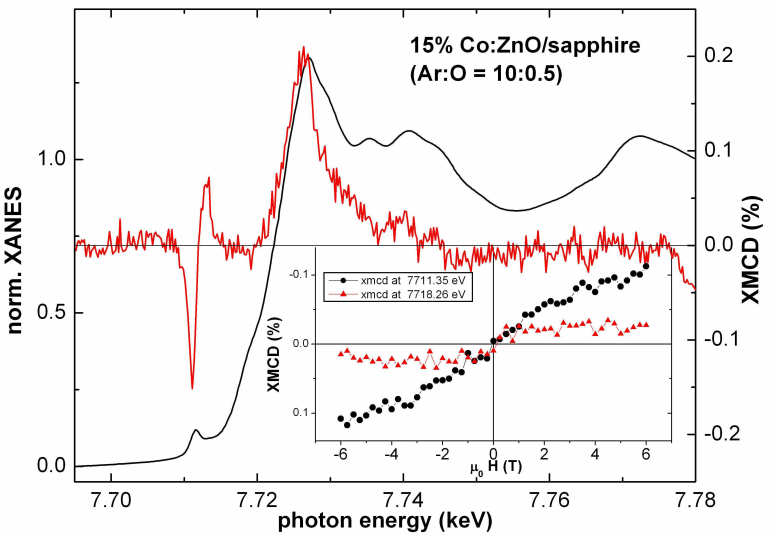

Figure 4: XANES and respective XMCD spectra of a SPM 15% Co:ZnO sample grown by RMS. The M(H) curves in the inset reveal PM at the pre-edge feature anda small XMCD signal of metallic Co.