ESRF	**Experiment title:** Probing the Mn incorporation, polarization and magnetism to evidence magneto-electric coupling in multiferroic $Ge_{1-x}Mn_xTe$	**Experiment number**: HC-3266
Beamline: ID 12	**Date of experiment**: from: 27.09.2017 to: 08.10.2017	**Date of report**: 03.03.2018
Shifts: 18	**Local contact(s)**: F. Wilhelm	*Received at ESRF:*

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Report:

The aim of the proposal was to study the structural, electric and magnetic properties of Co in ZnO and Mn in GaN by x-ray absorption near-edge spectroscopy (XANES), x-ray linear dichroism (XLD) and x-ray magnetic circular dichroism (XMCD) in external electric and magnetic fields to evidence magneto-electric coupling via changes in the anisotropic paramagnetism of both compounds. In a first step the structural quality of the Co:ZnO (Fig. 1) and Mn:GaN (Fig. 2) samples was checked on a local scale using XLD at room temperature. We consistently find a high structural anisotropy evidenced by a clear XLD signal typical for the wurtzite structure. We also assured that the used electrical top Au/(AlO_x) contact did not influence the XAS. Only at the Mn K-edge the signal to noise ratio is not ideal because of the low Mn concentration of

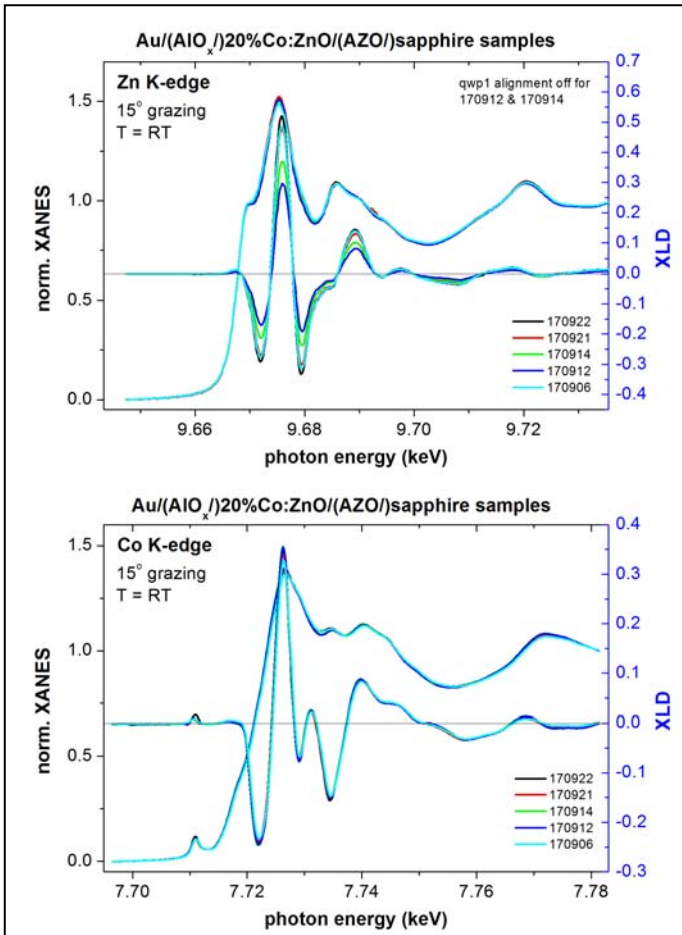

Figure 1: XANES/ XLD at the Zn (top) and Co (bottom) K-edges for various 20%Co:ZnO samples with top contacts.

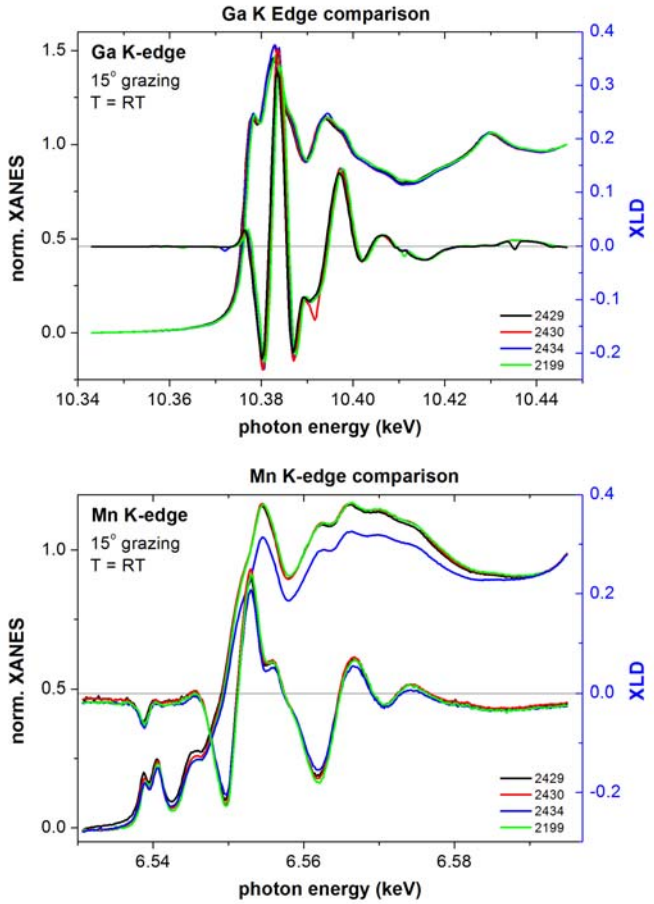

Figure 2: XANES/ XLD at the Ga (top) and Mn (bottom) K-edges for various 4%Mn:GaN samples with top contacts.

In a second step, the XANES of various samples have been measured under applied electric field. A representative outcome is shown in Figure 3 for Co:ZnO at the Co K-edge (top) and for Mn:GaN at the Mn K-edge (bottom). Unlike in previous experiments the XAS experienced various problems upon applying a voltage to the top contact. In the case of the Co:ZnO the asymmetric behavior can be interpreted as diode-like behavior. While for positive voltages the usual linear effect seems to be present while for negative voltages charging issues seem to dominate. In the case of Mn:GaN charging effects are present only at voltages higher than +/-300 V while at lower voltages a linear behavior with field is seen, however the shift of the XANES upon applying the voltage is not present. Similar to Co:ZnO the charging issues are more pronounced at negative voltages. The beamtime did not allow for a more in-depth analysis of this behavior.
Finally, the XMCD at the Co and Mn K-edges was recorded for one representative sample for each system. Figure 4 summarizes the outcome. For the Co XMCD of 20% Co:ZnO the XMCD at the pre-edge feature remains unaffected by applying a voltage, this is also corroborated by the XMCD(H) curve (Fig. 4, top). Only slight changes at the XMCD at the main absorption are visible which appear to be more pronounced for negative voltages – unfortunately time did not permit to also record the XMCD(H) for negative voltages. At the main absorption the XMCD(H) was also dominated by charging effects while at the pre-edge feature, where the absorption itself is much reduced, reliable XMCD(H) curves could be recorded.
For 4% Mn:GaN the situation is very different. As can be seen in Fig. 4 (bottom), there is no substantial XMCD at the main absorption which can be understood by the fact that Mn is virtually a spin-only paramagnetic impurity. However, at the pre-edge feature, there is a clear E-field induced change in the XMCD which is also corroborated by the XMCD(H) curves with and without applied voltage. While negative voltages reduce the XMCD, positive increases it. This can be interpreted as voltage-induced changes in the magnetic moment. Currently, theoretical calculations are underway to better understand this behavior. Obviously, the Mn states are situated close to the Fermi level in GaN and thus can be shifted by an electric field while for Co in ZnO the Co states are away from the Fermi level. In addition, the finite slope of the Co XMCD(H) curve corroborates the AFM coupling in this material which appears to be unaffected by the voltage. In contrast, the absence of a finite slope in the XMCD(H) curves for Mn in GaN corroborate that the next neighbor interaction in that system may be ferromagnetic; however, one has to consider the low amount of Mn pair in a sample with only 4% doping.

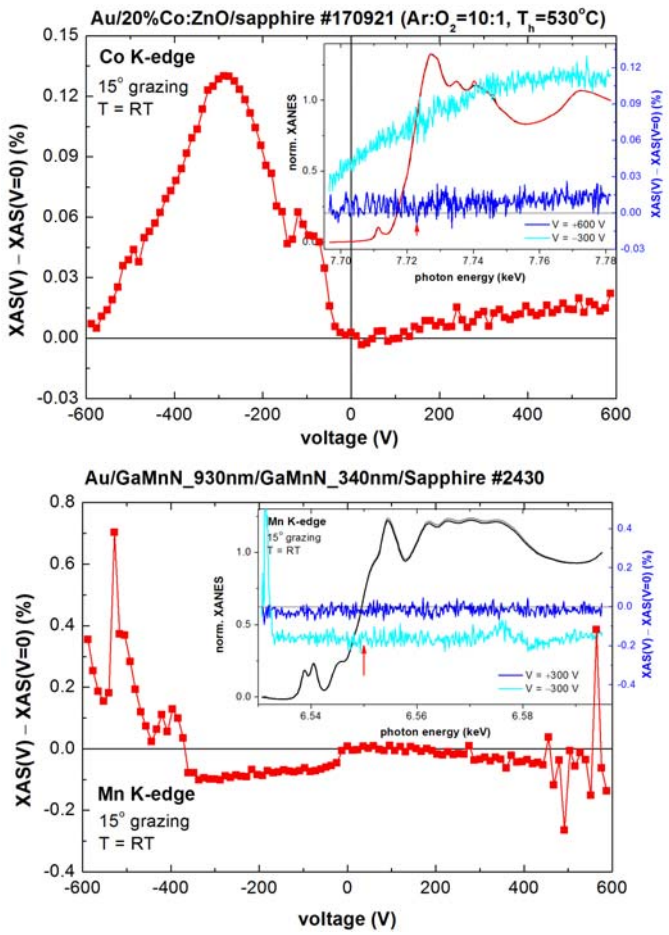

Figure 3: E field effect and field dependence (red arrows) at the Co K-edge (top) and the Mn K-edge (bottom).

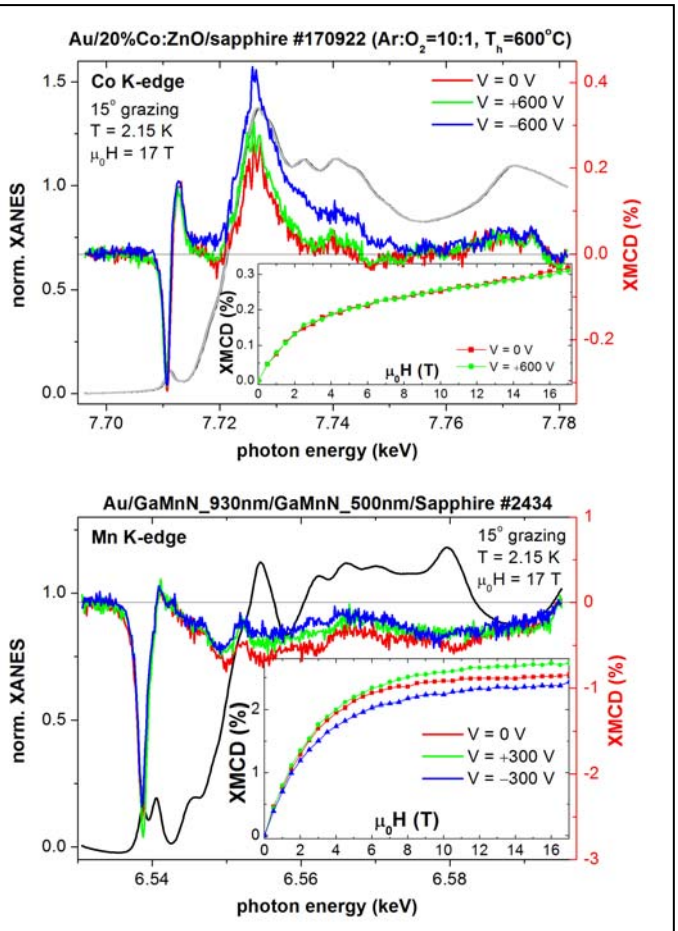

Figure 4: XANES, XMCD at 2K and 17T with and without E-field at the Co K-edge (top) and the Mn K-edge (bottom). The XMCD(H) has been recorded at the respective pre-