ESRF	**Experiment title:** Temperature dependence and detailed element specific hysteresis of the superdilute magnetic semiconductor Gd:GaN using the flipper magnet of ID 12	**Experiment number**: HE-2553
Beamline: ID 12	**Date of experiment**: from: 07.11.2007 to: 13.11.2007	**Date of report**: 12.02.2008
Shifts: 18	**Local contact(s)**: F. Wilhelm	*Received at ESRF:*

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Report:

First, we refined the element specific investigations on a MBE grown Gd:GaN sample which was studied earlier (A. Ney et al., APL **90**, 252515 (2007)). In Fig. 1 we summarize the x-ray magnetic circular dichroism (XMCD) measurements performed at the Ga K-edge. Both, the helicity of the light and the magnetization were reversed to rule out artefacts. We could detect a tiny XMCD signal of 0.04% for 15° grazing incidence of the light at low temperatures and high magnetic fields (Fig. 1a). However, the measurements under 15° grazing incidence suffer from residual x-ray linear dichroism (XLD) signatures originating from

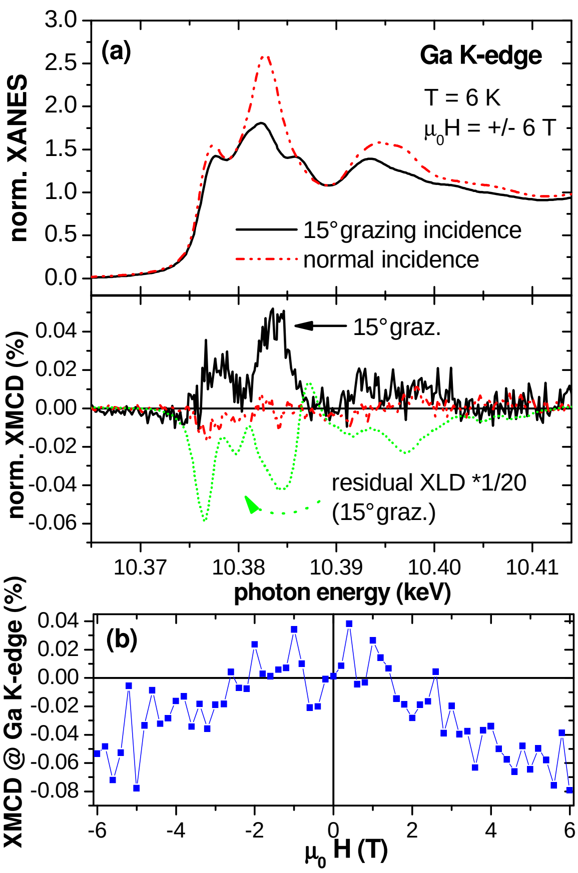

Figure 1: (a) XMCD spectra recorded under grazing and normal incidence for Gd:GaN at the Ga K-edge. Only a tiny signal is detectable dominated by residual XLD signatures. (b) The field dependence confirms the absence of magnetic polarization at the Ga sites.

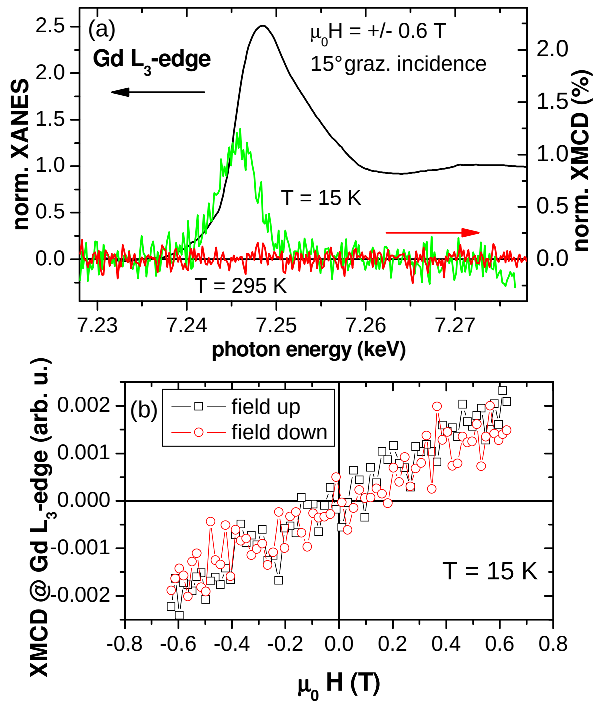

Figure 2: (a) Normalized XANES and XMCD spectrum at the Gd L3-edge of Gd:GaN using the flipper magnet. The dichroic signal is only detectable at very low temperatures. (b) The XMCD hysteresis recorded with high field resolution reveals neither significant remanence nor coercivity.

small linear components in the polarization which may affect the credibility of such a small XMCD signal. Therefore, we performed further XMCD measurements at the Ga K-edge for normal incidence of the light where the residual XLD signal is absent for normal incidence (Fig. 1a). For this experiment the XMCD signal is reduced to less than 0.01% with respect to the edge jump. Further, the element specific hysteresis (Fig. 1b) for grazing incidence corroborates that the XMCD signal measured under 15° grazing incidence is an artifact of the measurements since no conclusive field dependence is visible. Thus we conclude that no sizable magnetic polarization exists at the Ga sites of Gd:GaN. It should be noted that the Gd concentration in this sample was choosen such that the entire GaN matrix is expected to be polarized according to the previously proposed model (S. Dhar et al. PRL **94**, 037205 (2005)). In Fig. 2 we show XMCD (a) and hysteresis (b) measurements at the Gd L_3-edge of the same sample recorded using the flipper magnet. Whereas a small dichroic signal is visible at 15 K, it is absent at 100 K (not shown) and 300 K as well. The field dependence of the XMCD signal at 15 K was recorded with high field resolution and neither a remanence or a coercive field is visible. Thus we cannot provide any evidence of ferromagnetic properties for neither the Ga nor the Gd edges although for this Gd:GaN film SQUID measurements show a clear hysteresis at room temperature.

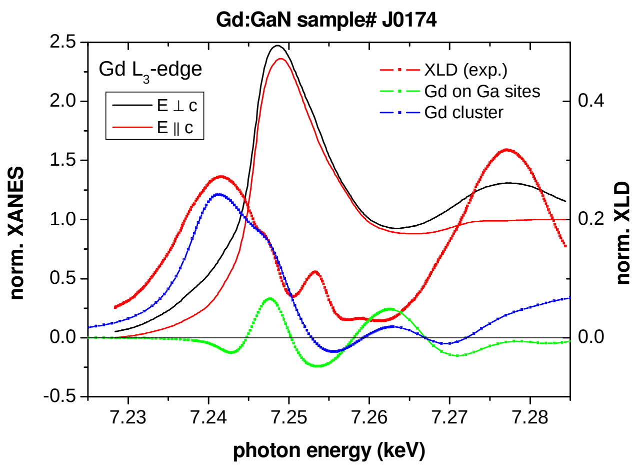

Figure 3: Normalized XLD specta at the Gd L_3-edge of a Gd: GaN sample with high Gd concentration together with simulations for substitutional Gd and Gd clusters. The XLD is very different from the sample with low Gd content clearly demonstrating the presence of clusters.

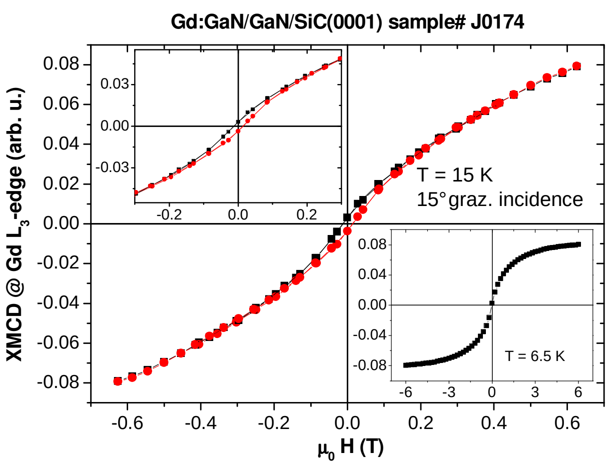

Figure 4: Element specific hysteresis at the Gd L_3-edge recorded at low temperatures for the clustered Gd:GaN sample.

In a second step we performed XLD and XMCD on an MBE-grown GaN sample with high Gd concentration (~2.9%). This sample is known to contain small phase seggregated clusters. Figure 3 shows the respective XLD measurements together with simulations using the FDMNES code (Y. Joly, PRB **63**, 125120 (2001)). Although the XLD spectrum is dramatically different from what can be measured in samples with lower Gd concentration (A. Ney et al., APL **90**, 252515 (2007)) The experiment can be fitted with resonable accuracy assuming Gd clusters, i.e. Gd atoms placed on both Ga and N sites along the c-axis. Based on the simulations also substitutional Gd, i.e. Gd on Ga sites cannot be excluded. The element specific hysteresis loop at the Gd L_3-edge for this sample is shown in Fig. 4 recorded using the flipper magnet (15 K) and the superconducting magnet (6.5 K). Whereas the superconducting magnet shows a paramagnetic-like field dependence, the higher field resolution of the flipper magnet reveals a small opening of the hysteresis loop indicative of ferromagnetic properties. However, the opening of the hysteresis vanishes above about 100 K, therefore the hysteresis is very likely to originate from tiny clusters with low ordering temperatures. We also measured XMCD spectra at the Ga K-edge for this sample at low temperatures (not shown) which give no evidence of magnetic polarization of the Ga sites. Therefore, the presence of the ferromagnetic Gd clusters does not influence the magnetic properties of the GaN host crystal.

In summary element specific investigations do not provide any evidence for ferromagnetic properties of the Ga and Gd sites in Gd:GaN for a wide Gd concentration range. The magnetic properties observed by integral SQUID measurements have to originate from elsewhere, e.g. the N sites or the O or H contamination which is always present in such samples.