	Experiment title: Study of ferromagnetism in wurtzite and cubic GaMnN semiconductors	**Experiment number**: HE-2007
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Report:

Boosted by the work of T. Dietl *et al*. [1], wide gap semiconductors like GaMnN might offer better possibility for Tc above room temperature. Dietl's calculations, based on the Zener model of ferromagnetism, predict that GaN doped with 5% Mn^{3+} and $3.5x10^{20}$ holes per cm^3 should exhibit the highest Curie temperature Tc (~420K). However, these calculations are unrealistic since today the highest hole concentration achieved in GaMnN is of the order of 10^{18} holes per cm^3. Today, more realistic models, bsed on Monte Carlo calculations or using an effective Heisenberg Hamiltonian, the T_C is predicted to be rather around ~50K for 7 at.% Mn in GaMnN (e.g. ref. [2]). All those calculations were mainly based on Mn 3+ valence state that corresponds to an electronic Mn $3d^5$ configuration. We have recently shown that the Mn electronic configuration is mainly 2+ ($3d^4$) for Mn incorporated in wurtzite GaN. The first goal of this proposal was to probe the 4p orbital magnetism via the XMCD at the Mn K-edge in ferromagnetic GaMnN wurtzite (n-type conductivity) and cubic (p-type conductivity) with similar Mn concentration.

High structural quality wurtzite and zinc-blende (cubic symmetry) GaMnN with a Mn concentration of ~2% have been grown by plasma-assisted molecular beam epitaxy (MBE). The wurtzite samples were grown on 1-mm-thick AlN-on-sapphire templates provided by NGK. The zinc-blende samples were grown on a SiC thick layer deposited by chemical vapor deposition on a Si substrate. Prior to the (Ga,Mn)N growth, a 300 nm GaN buffer layer was deposited by MBE. All the (Ga,Mn)N layers were grown to thickness 0.5 μm at a substrate temperature of 700°C under N-rich conditions, at a background pressure of 10^{-8} mbar, growth pressure of $2x10^{-5}$ mbar, and deposition rate of 0.25 μm/ hr. The Mn flux used during growth corresponds to a ~2–3 % Mn content, and was the same for all samples.

We have first measured the X-ray linear dichroism at the Mn K-edge in cubic and wurtziet $Ga_{1-x}Mn_xN$ in order to ensure the high structural quality. In Fig. 1, we have plotted the XLD signal recorded at the Mn K-edge and Ga K-edge for a $Ga_{1-x}Mn_xN$ thin film with x=2-3% Mn. The XLD was recorded using a quarter wave plate which allows to flip the linear polarization of the incident beam from horizontal to vertical at each energy point. The sample was measured at the 10° grazing incidence, in such a way that the horizontal linear polarization is parallel to the (a,b)-plane and the vertical polarization nearly perpendicular to the c-axis. The total fluorescence yield was collected via 8 photodiodes forming a large detection solid angle and mounted in backscattering geometry. We have observed the same Mn K-edge XLD signal (edge part) in shape and in amplitude (within 3%) as in similar wurtzite samples previously reported (HE-1894). ***The XLD experiments show that the Mn atoms are Ga substituted and do no reveal the presence of any secondary phases or metallic clusters.*** In the case of the cubic sample, we observed a rather small XLD signal that shows the presence of a secondary wurtzite phase. The XLD signal is about 10 times smaller compared to the wurtzite one. For cubic system, the XLD signal should be zero for dipolar transitions. The observed small XLD signal correspond to the XLD of wurtzite ones. The origin of the presence of 10% wurtzite phase in the cubic sample is up to now not clear, however it does not affect the isotropic xanes and xmcd spectra as we will see further. First, we can observe differences in the xanes pre-peak that consists mainly of a single peak in the case of the cubic sample contrary to the wurtzite one which consist of two pre-peaks. This difference reflects the change in the electronic configuration. In the case of the wurtzite sample, it has been demonstrated that the Mn electronic configuration is mainly 3+ ($3d^4$) [3]. Moreover the pre-edge signal is also more accentuated in the cubic sample compare the wurtzite one. Those differences reflect changes in the electronic configuration and/or band impurity level within the band gap above the Fermi level.

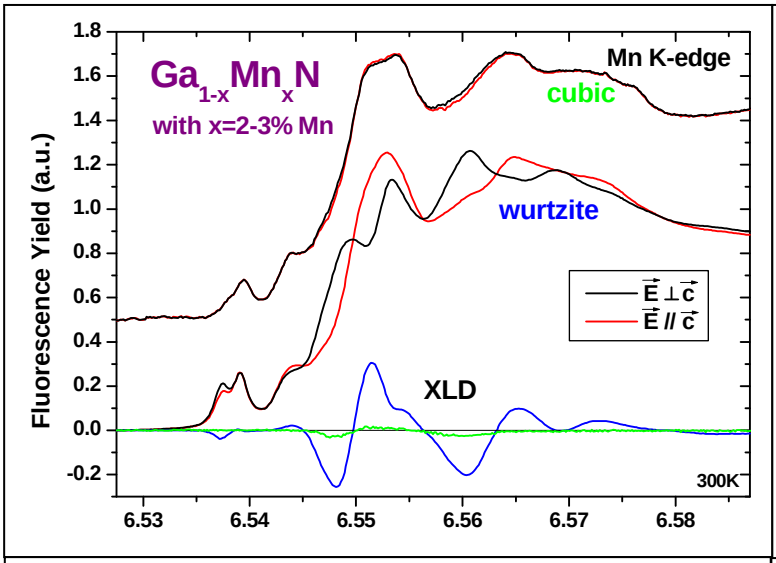

Fig.1 : X-ray Linear Dichroism recorded at the Mn K-edge at 300K in cubic (green colour) and wurtzite (blue colour) $Ga_{1-x}Mn_xN$ with a equivalent Mn concentration x=2-3%. The XANES spectra of both wurtzite and cubic samples have been normlized to an edge jump of unity for comparison.

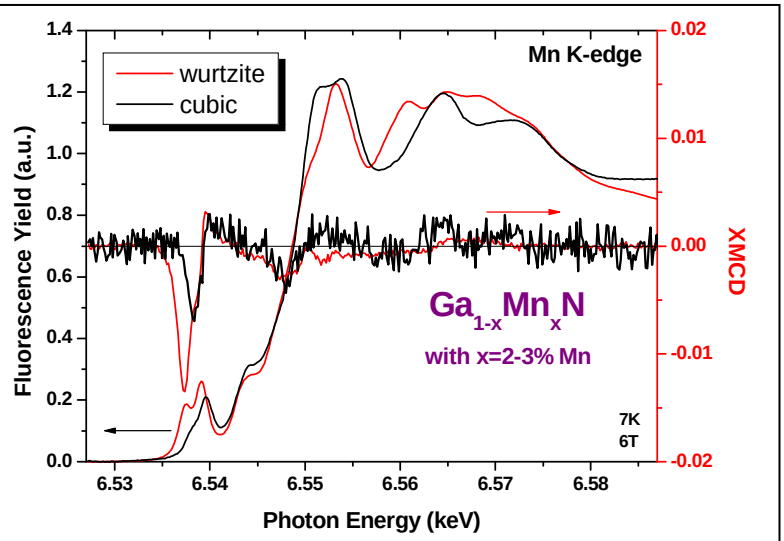

Fig.2 : X-ray Magnetic Circular Dichroism recorded at the Mn K-edge at 7K and under 6 Tesla external magnetic field in cubic (red colour) and wurtzite (black colour) $Ga_{1-x}Mn_xN$ with a equivalent Mn concentration x=2-3%.

In Fig. 2 are shown the X-ray Magnetic Circular Dichroism at the K-edge of Mn in $Ga_{1-x}Mn_xN$. A very intense XMCD signal (up to 1.4% with respect to the edge jump) is observed mainly at the first pre-peak of the XANES spectrum for the wurtzite sample. Since the integrated XMCD signal at the K-edge is proportional to the orbital polarization of the absorbing atom, our result clearly shows that the Mn atoms in (Ga,Mn)N carry an orbital magnetic moment. The observation of a sizeable orbital moment by XMCD is a strong argument in favor of the Mn^{3+} valence state. Indeed, in the case of pure Mn^{2+} where the 3*d* and 4*p* orbital moments are nearly zero, the XMCD signal is usually one order of magnitude smaller. In the case of the cubic sample, we observe also a clear XMCD signal that is however located at higher energy. The integral of this XMCD signal is ~5 times smaller than the one for the wurtzite sample. Moreover, it is interesting to observe that the XMCD signal (spectral shape and energy position) of the cubic ones matches

the XMCD signal at the shoulder of the wurtzite one. This is indeed a hint that the Mn electronic configuration of the wurtzite sample is not exactly $3d^4$ but mainly $3d^4$ within a mixed d^4-d^5 hybridized configuration [4]. Both wurtzite and cubic sample show however a paramagnetic behavior according to element specific magnetization curves done at the Mn K-edge at 7K.

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[3] E. Sarigiannidou, F. Wilhelm, E. Monroy, R. M. Galera, E. Bellet-Amalric, A. Rogalev, J. Goulon, J. Cibert, and H. Mariette, Phys. Rev. B74, 041306(R) (2006).
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