ESRF	**Experiment title:** **Fermi level tuning of ferromagnetism in a diluted magnetic semiconductor**	**Experiment number**: HE3138
Beamline: ID12	**Date of experiment**: from: 9/12/09 to: 15/12/09	**Date of report**: 12/8/10
Shifts: 17	**Local contact(s)**: Andrei Rogalev	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Kevin Edmonds*, Peter Wadley*

School of Physics and Astronomy, University of Nottingham, Nottingham NG7 2RD, United Kingdom

Gerrit van der Laan

Diamond Light Source, Didcot OX11 0DE, United Kingdom

Report:

Diluted magnetic semiconductors (DMSs) based on GaAs are of widespread interest as they offer a prototype system for exploring spin phenomena in technologically important III-V semiconductors. Importantly, their magnetic properties are determined by the concentration of spin-polarized charge carriers (holes). However, recent studies have provoked considerable debate as to whether these holes reside in a disordered valence band or a detached impurity band [1,2]. Here we present measurements of the As and Mn *K* edge x-ray magnetic circular dichroism (XMCD) in a series of (Ga,Mn)As and (In,Ga,Mn)As films, which shed new light on the element-resolved character of the states at the Fermi energy in these materials.

The (Ga,Mn)As and (In,Ga,Mn)As films were grown by molecular beam epitaxy at the University of Nottingham, on GaAs(001) and InP(001) substrates respectively. In order to prevent the substrate from dominating the As *K* absorption signal, the (Ga,Mn)As films were grown on a 200nm thick AlAs buffer layer, which was subsequently removed by chemical etching to leave a free-standing layer. Some of the films were subjected to a low-temperature annealing procedure in order to remove hole-compensating Mn interstitial defects from the layers. Furthermore, in order to introduce additional compensation, one of the samples was H^+ ion-implanted with a dose equivalent to the substitutional Mn concentration. This was found to fully suppress the conductivity and ferromagnetism in the film. The Mn and As *K* edge x-ray absorption and XMCD measurements were obtained in fluorescence yield mode on line ID12 of ESRF, at a sample temperature T=7K and under a magnetic field B=6T.

Figure 1 shows the Mn *K* edge absorption and XMCD spectra from a 5% Mn doped (Ga,Mn)As film before and after H^+ ion implantation, as well as a 1% Mn doped layer. The XMCD spectra consist of three distinct peaks, two of which occur in the vicinity of the x-ray absorption pre-edge structure, and the other of which is coincident with the main absorption edge. All three peaks vary significantly in intensity from sample

to sample, with the lowest energy peak showing the largest variation. The XMCD is very weak in the ion implanted film, even at T=7K and B=6T, indicating that, in the absence of hole-mediated ferromagnetic interactions, the interaction between the Mn ions is antiferromagnetic. The most dramatic feature of Fig. 1 is that the XMCD is greatly enhanced for the 1% doped layer compared to the 5% doped layers.

Figure 2(a) shows the As *K* edge absorption and XMCD spectrum from the 5% Mn doped layer. A sizeable XMCD is observed, around a factor of three larger than in our previous study [3], which is ascribed to the higher sample quality and reduced density of compensating interstitial defects. The XMCD is centered at an energy just lower than the absorption edge. A comparison with calculated As *K* absorption spectra obtained using the FDMNES code [4], presented in Fig. 2(b), shows that this region of the spectrum is sensitive to small changes in the Fermi energy. This indicates that the As *K* edge XMCD is associated with transitions to valence band states that become unoccupied due to Mn acceptor doping.

The integrated XMCD intensity can be related to the orbital magnetic moment of the As 4*p* states on a per atom basis by applying the orbital moment sum rule. The obtained orbital moment m_{orb}^{4p} per As ion is plotted in Fig. 3 for six different (Ga,Mn)As and (In,Ga,Mn)As films, as a function of the saturation magnetization M_S which is a measure of the concentration of magnetically active Mn ions. It is observed that m_{orb}^{4p} increases linearly with M_S for values above ~15 emu/cm^3 (equivalent to a substitutional Mn concentration of around 2%), reaching values in excess of $0.003\mu_B$ /As in the highest doped sample studied. The hole density for this sample is estimated from high field Hall effect measurements to be around $8\text{x}10^{20}\text{cm}^{-3}$, which points to an As 4*p* orbital moment per valence band hole of around $0.1\mu_B$. In addition, from the sign of the XMCD we determine that the As 4*p* orbital moment is of opposite sign to the Mn 3*d* local magnetic moments, in agreement with our previous study [3].

For lower Mn concentrations, the As *K* edge XMCD is below the noise level. Together with the dramatic enhancement of the Mn *K* edge XMCD for the lowest doped sample, this strongly suggests that the holes are built mostly from the 4*p* states of the impurity ions at low concentrations, but take on the character of the host as the Mn concentration increases. This apparent cross-over of the character of the hole states has important implications for the physics of ferromagnetism in these systems and warrants further investigation, in particular to determine its relation to the metal-to-insulator transition observed with increasing Mn doping. Furthermore, alloying the host semiconductor with a wider bandgap material such as AlAs or GaP may be expected to shift the cross-over to higher Mn concentrations, so that the dependence of the character of the hole states on the impurity bandwidth may be accurately determined.

References:

[1] K Burch *et al.*, Phys. Rev. Lett. **97**, 087208 (2006); K Ando *et al.*, Phys. Rev. Lett. **100**, 067204 (2008).
[2] T Jungwirth, J Sinova, AH MacDonald *et al.*, Phys. Rev. B **76**, 125206 (2007).
[3] AA Freeman, KW Edmonds, G van der Laan *et al.*, Phys. Rev. B **77**, 073304 (2008).
[4] Y Joly, Phys. Rev. B **63**, 125120 (2001).
[5] BT Thole, P. Carra, F Sette, and G van der Laan, Phys. Rev. Lett. **68**, 1943 (1992).

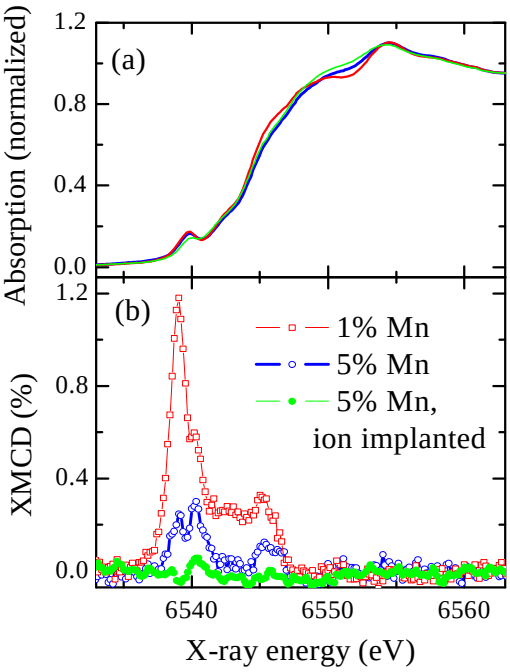

Fig. 1(a) Mn K edge absorption spectrum and (b) XMCD for (Ga,Mn)As films with 1% Mn doping (red), 5% Mn doping before (blue) and after (green) H ion implantation.

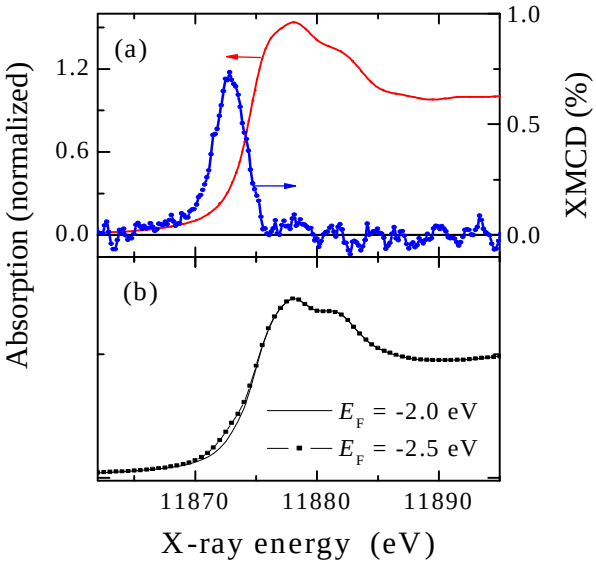

Fig. 2(a) As K edge absorption (red curve, left axis) and XMCD (blue curve, right axis) for a (Ga,Mn)As layer with 5% Mn doping. (b) Calculated As K absorption spectra obtained using FDMNES [4].

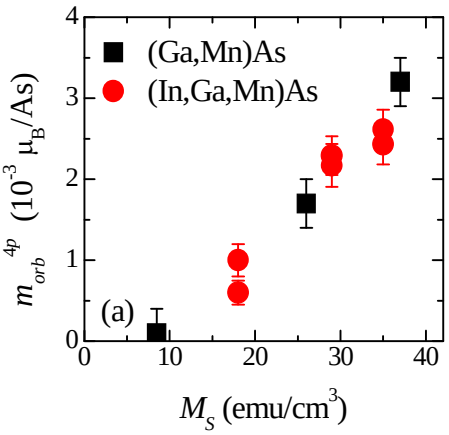

Fig. 3. As 4p orbital magnetic moment per ion, obtained by sum rule analysis of the As K edge spectra, versus the saturation magnetization obtained by SQUID magnetometry.