ESRF	**Experiment title:** Orbital polarization of itinerant and localized moments in a ferromagnetic semiconductor	**Experiment number**: HE-2386
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Report:

The (III,Mn)V magnetic semiconductors are model systems for investigating localized moment ferromagnetism [1,2] and new phenomena in spintronics [3,4]. In these systems, ferromagnetic interaction between the d^5 moments of the dilute Mn^{2+} ions is mediated by spin-polarized valence band holes [5]. The strong spin-orbit interaction felt by the holes leads to large anisotropies in the magnetic [6] and magnetotunnelling [3] properties, which are closely correlated with the symmetry and strain of the lattice. Understanding these phenomena, and their relationship to the hole orbital polarization, is essential for harnessing such effects in a new generation of spintronic sensors and memory elements [7]. Mn $L_{2,3}$ edge XMCD has been widely used to determine the magnetic and electronic properties of the localized Mn moment, while bulk magnetometry and magneto-optical measurements contain contributions from both localized and itinerant moments. However, until now there have been no experimental studies which quantitatively and separately capture the magnetism and anisotropy of the hole subsystem.

In this report, we exploit *K*-edge XMCD to directly probe the 4*p* valence states which are crucial for ferromagnetism in the (III,Mn)As family of magnetic semiconductors. Since *K*-edge absorption involves excitations from the 1*s* core level, a significant XMCD will occur only if there is an orbital polarization of the valence states that are probed. We present the first direct determination of the As and Ga 4*p* orbital magnetic moments in III-V ferromagnetic semiconductors.

The penetration depth of x-rays at the Ga and As *K*-edges is several tens of microns, which is much larger than the thickness of typical (Ga,Mn)As films. Therefore, Ga and As absorption signals from the substrate must be avoided. We investigated two different samples: (a) a 1μm thick (Ga,Mn)As film grown on an AlAs buffer layer on GaAs(001), released from the substrate by etching the AlAs layer, and re-mounted on sapphire; (b) a 0.5μm thick (In,Ga,Mn)As film on InP(001), which is nearly lattice-matched to the substrate by varying the In content in the film [8]. X-ray diffraction measurements show that the latter film is under small compressive strain. The films were grown by low-temperature (~200-250^{o}C) molecular beam epitaxy, and the nominal Mn concentration (estimated from the Mn flux during growth) was around 8%. SQUID magnetometry measurements show that the (Ga,Mn)As and (In,Ga,Mn)As films are ferromagnetic with T_C of 54 K and 21 K, respectively.

The XMCD measurements were performed on ID12 of the European Synchrotron Radiation Facility (ESRF) at Grenoble. Ga, As and Mn *K*-edge absorption spectra were obtained from total fluorescence yield measurements using 98% circularly polarized x-rays, and the XMCD was obtained from the difference in absorption for parallel and antiparallel alignments of the x-ray polarization vector with respect to an external magnetic field. To avoid experimental artefacts, the external magnetic field direction and the x-ray helicity were alternately flipped. The measurements were performed at a sample temperature of 10 K, under a magnetic field of ±2 T, which was either perpendicular or nearly parallel (~15^o) to the sample surface.

Figure 1(a-c) show the As *K*-edge absorption spectrum from the (Ga,Mn)As film, and the Ga and As *K*-edge absorption spectra from the (In,Ga,Mn)As film. A clear dichroism can be observed at the onset of the As absorption edge, indicating a polarization of the As 4*p* states at the valence band edge. The position and shape of the XMCD spectrum is similar for the two films, and also qualitatively similar to the main feature observed in As L_3 XMCD [9]. We also observe a substantial anisotropy of the As XMCD for the strained (In,Ga,Mn)As film, with a larger signal for grazing incidence than for normal incidence. At the Ga edge the XMCD is much weaker, and is scarcely visible above the noise level. By applying the XMCD orbital moment sum rule to the spectra, we obtain an As 4*p* orbital moment of around 10^{-3} μ_B per As ion, or around 0.1-0.2 μ_B per valence band hole.

Figure 2 shows Mn *K*-edge x-ray absorption and XMCD spectra from the (Ga,Mn)As and (In,Ga,Mn)As films. The main part of the Mn XMCD consists of a double peak structure with a splitting of ~1.4eV, centered around the pre-edge peak in the x-ray absorption spectrum. The Mn and As *K*-edge XMCD signals have the same sign, and are of comparable magnitude. We ascribe the features in the Mn *K* pre-edge XMCD to a mixing between Mn 3*d* and 4*p* states, where the 3*d* and 4*p* orbital moments are antiparallel.

In summary, we have used XMCD to directly determine the magnitude and character of the valence band orbital magnetic moments in (III,Mn)As ferromagnetic semiconductors. A distinct dichroism is observed at the As K absorption edge, yielding an As 4p orbital magnetic moment of around -0.1μ_B per valence hole, which is strongly influenced by strain. The dichroism at the Ga K edge is much weaker. The K edge XMCD signals for Mn and As both have positive sign, which indicates the important contribution of Mn 4p states to the Mn K edge spectra.

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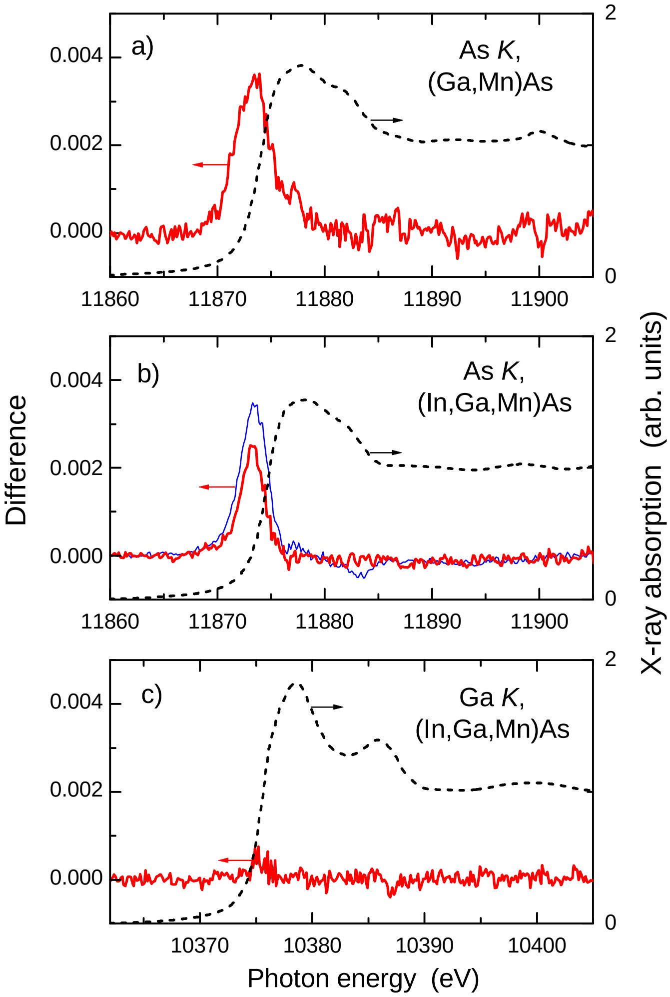

FIG. 1. XMCD spectra (full lines, left axes) and x-ray absorption spectra (dotted lines, right axes) for (a) As *K* edge of (Ga,Mn)As; (b) As *K* edge of (In,Ga,Mn)As; (c) Ga *K* edge of (In,Ga,Mn)As. Thick red lines give the XMCD for normal incidence, and the thin blue line in (b) gives the XMCD for grazing incidence.

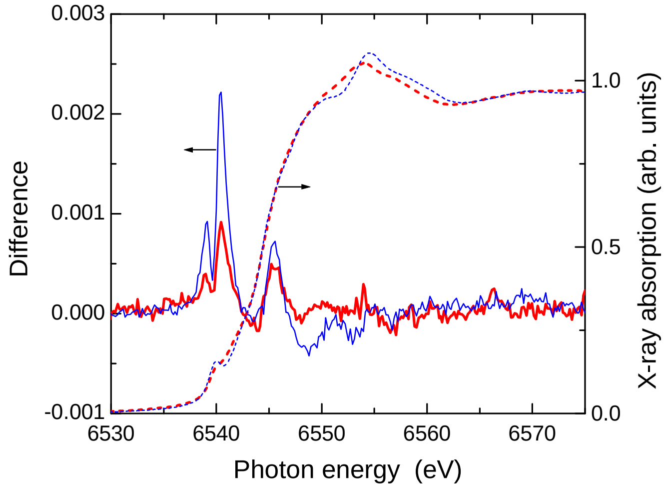

FIG. 2. Mn *K* edge XMCD spectra (full lines, left axis) and x-ray absorption spectra (dotted lines, right axis) for (Ga,Mn)As (thin blue lines) and (In,Ga,Mn)As (thick red lines).