Element-resolved Spin Configuration in the Ferromagnetic Semiconductor (Ga,Mn)As

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Introduction

Semiconductor materials doped with magnetic ions can display both ferromagnetism and semiconducting electronic properties and are thus of intense interest for creating a new class of devices that combine bandgap engineering with control of electron spin [1]. However, the origin of the ferromagnetic coupling between the widely separated magnetic impurities is not fully understood. Principal among this class of materials has been Mn-doped GaAs, due to its status as the only III-V based magnetic semiconductor and its excellent magnetic properties [2, 3]. Consequently, this material has been extensively studied both structurally and electronically, and there is a consensus that it truly represents an intrinsic combination of ferromagnetism and semiconducting band structure, as opposed to a minority phase. Therefore, by examining the origin of ferromagnetism in this material, we gain insight into the mechanism of ordering in general.

Several recent band structure calculations based on the assumption of Mn substitution into Ga sites suggest that the Mn ions are coupled via a double exchange mechanism mediated by unoccupied valence band states (holes) primarily derived from the As 4sp atomic levels [4-6]. In this model, the As is predicted to have a small, induced spin polarization antiparallel to that of the neighboring Mn ions, while Ga has an even smaller polarization parallel to the Mn. Therefore, by measuring these induced magnetic moments, we are able to test current models of ferromagnetic ordering in magnetic semiconductors.

Methods and Materials

We have recently used soft x-ray magnetic circular dichroism (XMCD) at the Mn, As, and Ga L edges to detect and determine the relative orientations of these predicted induced moments [7]. The L absorption edges probe the 2p→3d and 2p→4s transitions. Thus, by exciting with circularly polarized radiation and measuring the differential absorption between parallel and antiparallel photon helicity and sample magnetization, we obtain the magnetic structure of the unoccupied Mn 3d and Ga and As 4s states. This differential absorption is related to the average magnetic moment of the 3d or 4s orbitals. Therefore these measurements yield the *element-specific* net spin orientations for all three elements.

Epitaxial films of $Ga_{1-x}Mn_xAs$ with x = 0.039, 0.049, 0.059, 0.07, and 0.081 were grown on GaAs(100) wafers by molecular beam epitaxy (MBE) in an As-rich environment. The samples were grown at 250ºC and were not annealed after deposition. The XMCD measurements were done at the soft x-ray beamline station 4-ID-C at the APS. This beamline uses a circular polarized undulator that provides >96% left- or right-circular polarization in the energy range from 500 to 3000 eV [8]. Samples were cooled to ~30K, and x-ray absorption and dichroism measured in total electron yield with the beam incident at 20° above the film plane. Dichroism scans were taken at fixed photon helicity by reversing the magnetization of the sample using an in-vacuum electromagnet and measuring in remanence at each energy.

Results

Figure 1, is a plot of the x-ray absorption spectra (blue curves) and XMCD (green curves) at the Mn, As, and Ga L edges for the x = 0.07 sample. The Mn XAS shows a two-phase structure, with a nonmagnetic Mn^{2+} multiplet (features B, C, D, E) that is associated with a surface oxide and with a magnetic component (feature A) that is the ferromagnetic (Ga,Mn)As phase. All of the samples showed Mn dichroism with an identical lineshape, indicating ferromagnetic ordering of the Mn. In addition, dichroism signals were detected at the Ga L_3 edge (1116 eV) for x = 0.049 and above. At the As L_3 edge (1324 eV), dichroism was observed for the x = 0.07 and 0.081 samples. These dichroism signals indicate induced polarization of the As and Ga.

Figure 2, shows hysteresis loops and temperature dependence data taken at the Mn and Ga dichroism peaks for the x = 0.049 sample. The Ga moment shows the same switching behavior and curie temperature. This indicates that the Ga moment is clearly associated with the ferromagnetic phase of (Ga,Mn)As.

Discussion

The sign of the As dichroism is the same as that of the Mn 3d dichroism. However, analysis of the dipole selection rules shows that the sign of the dichroism for p→s transitions is opposite that for p→d transitions [9]. Therefore, this indicates that the As 4s moments are

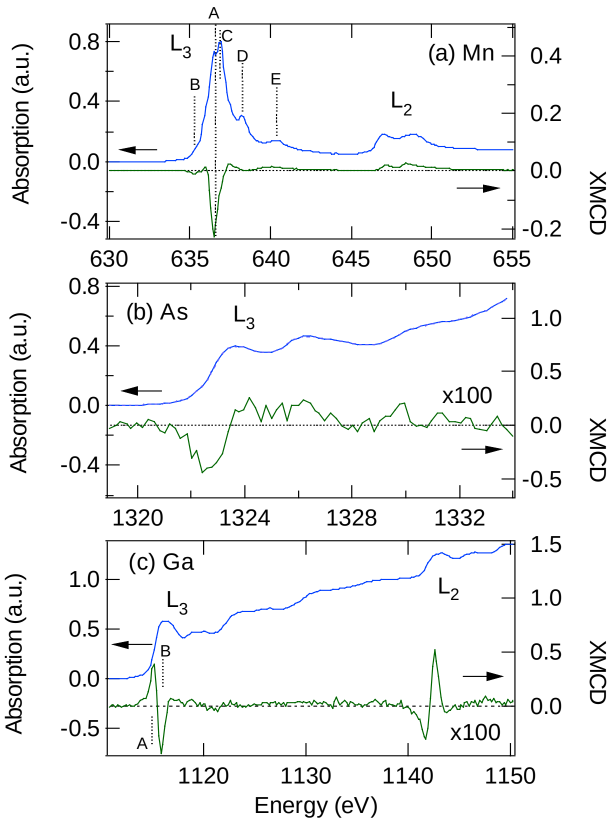

FIG. 1. XAS and XMCD data taken for $Ga_{0.93}Mn_{0.07}As$ at 30-40K at the Mn (a), As (b), and Ga (c) L edges

antiparallel to the Mn. The bipolar nature of the Ga signal makes a determination of the sign of the Ga moment more ambiguous. However, we note that both the initial dichroism as well as the integrated L_3 curve are opposite in sign to the As dichroism. Since the magnetic moment is proportional to the integrated dichroism, we assign the Ga as being antiparallel to the As. Therefore, the relative spin orientations, according to the 4s moments, are as predicted.

A quantitative estimate of the total As and Ga moments is not possible from these data, because a significant portion of those moments are in the 4p states, which are not probed by the L edges. However, our results are at least consistent with the calculated moments. The band structure calculations predict a significantly higher moment on As than on Ga. Analysis of the areas under the respective L_3 XMCD curves shows that the Ga 4s moment is probably several times smaller than the As 4s moment.

In summary, we have detected, for the first time, induced host moments in a ferromagnetic semiconductor, and we have determined the relative spin orientations of all the elements in the material. The results are consistent with recent predictions based on band structure calculations,

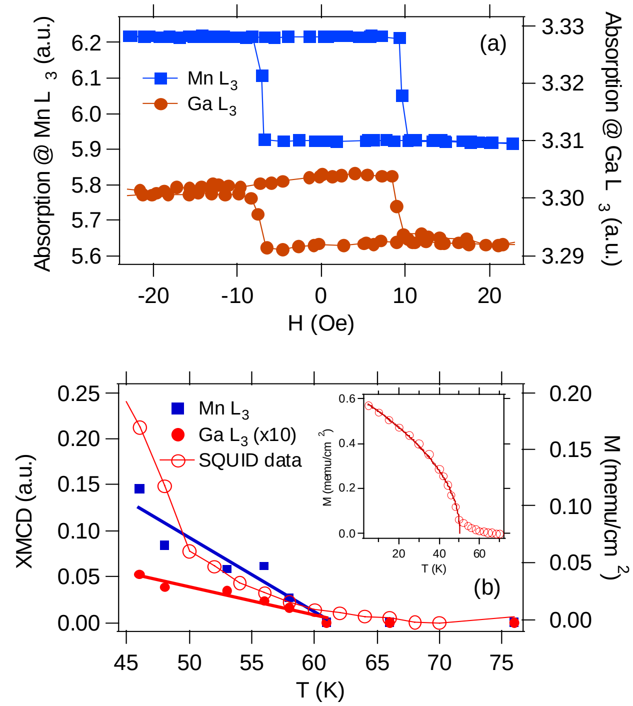

FIG. 2. (a) Hysteresis at 45K of the Mn and Ga L_3 dichroism peak intensities for $Ga_{0.951}Mn_{0.049}As$. (b) Temperature dependence of the dichroism for the same sample. Open circles show the total magnetization of the same sample obtained by SQUID magnetometry.

and they lend support to the theory of carrier-mediated double exchange as the origin for ferromagnetism in such systems.

Acknowledgments

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