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Published in:
Journal of Applied Physics

Link to article, DOI:
10.1063/1.2712165

Publication date:
2007

Document Version
Publisher's PDF, also known as Version of record

Link back to DTU Orbit

Citation (APA):

DTU Library
Technical Information Center of Denmark

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Element specific investigation of ultrathin Co$_2$MnGa/GaAs heterostructures

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(Received 30 October 2006; accepted 18 December 2006; published online 3 May 2007)

We have used x-ray magnetic circular dichroism to study the element specific magnetic properties of ultrathin films of the Heusler alloy Co$_2$MnGa at room temperature. Nine films were grown by molecular beam epitaxy on GaAs substrates and engineered to vary in stoichiometry as Co$_{1.86}$Mn$_{0.99}$Ga$_{1}$, Co$_{1.95}$Mn$_{0.98}$Ga$_{1}$, and Co$_{1.97}$Mn$_{0.96}$Ga$_{1}$, with thicknesses of 5.3, 7.6, and 9.7 nm, and were capped with Al to prevent oxidization. Sum rule analysis revealed magnetic moments significantly lower than the values predicted theoretically, especially in the case of the Mn total magnetic moment. The results do not support the hypothesis that a large magnetic moment is transferred from Co to Mn, and may suggest that diffusion and reacted layers at ultrathin thicknesses can effect the establishment of a full Heusler structure. © 2007 American Institute of Physics.

[DOI: 10.1063/1.2712165]

I. INTRODUCTION

The fabrication of Heusler alloys, which are ferromagnetic alloys with nonferromagnetic constituents, was first reported in 1903. From the outset, these alloys created a great deal of interest in both experimental and theoretical fields. Recently, Heuslers have once more come to the forefront of research as their half-metallic properties favor both the spin polarized transport and integration with semiconductor structures required for spintronic devices.

One such Heusler/semiconductor heterostructure is the combination Co$_2$MnGa grown on GaAs(100). Co$_2$MnGa is a full Heusler of the L$2_1$ structure, whose combination with GaAs appears possible due to a fairly low lattice mismatch ($<2\%$), and has proved promising in previous investigations by our collaborators at Toshiba Europe. Despite considerable interest in integrating ultrathin Heusler films with semiconductors such as GaAs for the development of next generation spintronic devices, only a limited number of studies on the element specific magnetic moments of such structures have taken place. Grabis et al. found that the reduced magnetization of Co$_2$MnGe thin films is due to a reduction of the Mn moments. Our detailed x-ray magnetic circular dichroism (XMCD) studies of ultrathin Co$_2$Mn$_x$Ga$_{1-x}$ films, presented here, show that while the Mn moments are comparable to those of Co, they are much smaller than the theoretically predicted values. Our study suggests that the structure, composition, and, thus, the magnetic properties of the ultrathin Heusler films might be distinctly different from the bulk materials due to interface, strain, and possible interdiffusion.

II. EXPERIMENT

The samples were grown at the Technical University of Denmark by molecular beam epitaxy. After heating the epi-ready GaAs substrate to 650 °C to desorb oxygen from the surface, the substrate was allowed to cool down to 200 °C before the evaporation of the alloy films was carried out at a pressure of $5 \times 10^{-9}$ mbar. Growth was manipulated to achieve three stoichiometries: Co$_{1.86}$Mn$_{0.99}$Ga$_{1}$, Co$_{1.95}$Mn$_{0.98}$Ga$_{1}$, and Co$_{1.97}$Mn$_{0.96}$Ga$_{1}$, representing Co/Mn ratios of 1.88, 1.99, and 2.05, respectively, and the thicknesses of 5.3, 7.6, and 9.7 nm achieved through the use of a shutter. Oxidization during transfer was prevented by incorporation of a 1.5–1.7 nm Al capping layer. Full details of these processes can be found in Ref. 13.

XMCD measurements were performed at room temperature on station 1.1 of the Synchrotron Radiation Source (SRS) at Daresbury Laboratory using the portable octopole magnetic system (POMS), able to supply a field of 0.8 T in an arbitrary direction. The experimental setup is schematically shown in Fig. 1. X-ray absorption spectra (XAS) at the Mn and Co L$_{2,3}$ edges were obtained by recording the sample current in total electron yield (TEY) mode as a function of incident photon energy. The I$^+$ and I$^-$ XAS spectra,
corresponding to the photon helicity parallel and antiparallel, respectively, to the external field, were obtained by reversing the direction of the applied magnetic field at each point of the spectrum. The difference between the $I^+$ and $I^-$ spectra gives the XMCD spectrum.

At the Mn and Co $L_{2,3}$ edges, 2$p$ core electrons are excited into unoccupied 3$d$ states, allowing us to directly investigate the magnetically polarized 3$d$ band. XMCD sum rule analysis makes it possible to obtain the magnetic moments of both the Co and Mn components on a per atom basis.\textsuperscript{15,16} Our data were analyzed by the technique of Chen et al.\textsuperscript{16} using the standard equations; as described previously,\textsuperscript{17} the spin and orbital magnetic moments per atom are given as

$$m_{\text{orb}} = -4q n_p/3r P,$$

$$m_{\text{spin}} = (4q - 6p) n_p/r P.$$  

The number of 3$d$ holes, $n_p$, was taken to be 2.2 for Co and 4.5 for Mn based on the work of Schmalhorst et al.\textsuperscript{18} and the degree of x-ray polarization, $P$, to be 0.70. It is well known that in the case of Mn, the sum rules require a correction, since they do not take into account the $jj$ mixing,\textsuperscript{i.e.}, the mixing of the 2$p_{3/2}$ and 2$p_{1/2}$ core levels caused by covalence interactions. In a one-electron band structure model, the correction factor is equal to 1, but for a completely localized system, the expected correction factor is 1.47.\textsuperscript{20} A localized Mn system would show a narrow linewidth and a distinctive multiplet structure represented by two pronounced structures in the $L_3$ tail and a distinctive $L_2$ split peak such as seen in ultrathin Mn films\textsuperscript{21} or the ferromagnetic semiconductor (Ga,Mn)As.\textsuperscript{20} Based on the interpretation of the XAS line shapes of the Heusler alloys, which appear indicative of a fairly itinerant Mn system, the correction factor for this effect might be expected to be between 1 and 1.1, and would thus fall within the 10% error bar of the XMCD.

### III. RESULTS AND ANALYSIS

The XMCD of each of the nine samples were measured three times for each element (see Fig. 2) and the sum rules were applied to the resultant spectra. The average spin and orbital moments for each sample were then summed to give $m_{\text{tot}}$, a quantity directly related to the total moment per atom; these results are listed in Table I.
depth of ~5 nm (Ref. 24) results in the capping layer impinging on the thickness which may be viewed. This suggests that the more Heusler-like results of the 5.3 nm thickness of the higher ratio samples, Co/Mn are 2.0 and 2.1, implying that the alloy film does not have uniform magnetic properties. At this proximity to the interface, it is expected that such problems may be related to the diffusion and the reacted layers emanating from the interface, preventing the establishment of a true full Heusler structure.

IV. CONCLUSIONS

We have used XMCD to measure the element specific magnetic moments of three different thicknesses of the Heusler film CoMn$_x$Ga grown on GaAs(001), where the film stoichiometry was varied through $x$ and $y$ to assume the ratios of Co/Mn of 1.88, 1.99, and 2.05. The results did not demonstrate the expected large transfer of moment from the Co to the Mn site, but instead showed a deficit in the moment values. The thickness dependence of the results appears to point to a certain degree of magnetic nonuniformity through the alloy structure, with the Al capping layer and limited escape depth of the electron yield method preventing analysis down to greater depths. It remains a challenge to improve the growth and interface of Heusler alloy/semiconductor heterostructures for spintronics applications.

ACKNOWLEDGMENTS

This work was carried out with the aid of an EPSRC/Daresbury Laboratory CASE studentship and the Danish Agency for Science Technology and Innovation (FNU and FTP). We would like to thank N. D. Telling and N. R. S. Farley at SRS, Daresbury Laboratory, for their assistance with the POMS system and our collaborators S. Holmes and A. Husmann at Toshiba Europe.


TABLE I. The sum of orbital and spin moments (with error bars) in $\mu_B$/atom for the Co and Mn components as obtained from XMCD measurements on three different film structures of Co$_x$Mn$_y$Ga (indicated by the $x/y$ ratio) with three different thicknesses.

<table>
<thead>
<tr>
<th>Thickness (nm)</th>
<th>Co/Mn=1.88</th>
<th>Co/Mn=1.99</th>
<th>Co/Mn=2.05</th>
</tr>
</thead>
<tbody>
<tr>
<td>Co$<em>{m</em>{tot}}$</td>
<td>0.283 ±0.026</td>
<td>0.534 ±0.050</td>
<td>0.438 ±0.043</td>
</tr>
<tr>
<td>Mn$<em>{m</em>{tot}}$</td>
<td>0.206 ±0.019</td>
<td>0.175 ±0.016</td>
<td>0.177 ±0.017</td>
</tr>
<tr>
<td>Co$<em>{m</em>{tot}}$</td>
<td>7.6</td>
<td>0.724 ±0.038</td>
<td>0.438 ±0.049</td>
</tr>
<tr>
<td>Mn$<em>{m</em>{tot}}$</td>
<td>0.434 ±0.033</td>
<td>0.305 ±0.060</td>
<td>0.300 ±0.060</td>
</tr>
<tr>
<td>Co$<em>{m</em>{tot}}$</td>
<td>0.7</td>
<td>0.065 ±0.028</td>
<td>0.521 ±0.034</td>
</tr>
<tr>
<td>Mn$<em>{m</em>{tot}}$</td>
<td>0.276 ±0.026</td>
<td>0.283 ±0.026</td>
<td>0.283 ±0.026</td>
</tr>
<tr>
<td>Co$<em>{m</em>{tot}}$</td>
<td>9.3</td>
<td>0.438 ±0.028</td>
<td>0.467 ±0.028</td>
</tr>
<tr>
<td>Mn$<em>{m</em>{tot}}$</td>
<td>0.521 ±0.026</td>
<td>0.304 ±0.026</td>
<td>0.304 ±0.026</td>
</tr>
<tr>
<td>Co$<em>{m</em>{tot}}$</td>
<td>0.534 ±0.026</td>
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</tr>
</tbody>
</table>

*Reference 9.*