

# Temperature dependence of the interface moments in Co<sub>2</sub>MnSi thin films

N. D. Telling<sup>1</sup>, P. S. Keatley<sup>2</sup>, L. R. Shelford<sup>2</sup>, E. Arenholz<sup>3</sup>, G. van der Laan<sup>1,4</sup>, R. J. Hicken<sup>2</sup>,  
Y. Sakuraba<sup>5</sup>, M. Oogane<sup>5</sup>, Y. Ando<sup>5</sup>, and T. Miyazaki<sup>5</sup>

<sup>1</sup>*Magnetic Spectroscopy Group, STFC Daresbury Laboratory, Warrington WA4 4AD, United Kingdom*

<sup>2</sup>*School of Physics, University of Exeter, Stocker Road, Exeter EX4 4QL, United Kingdom*

<sup>3</sup>*Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*

<sup>4</sup>*Diamond Light Source, Chilton, Didcot OX11 0DE, United Kingdom*

<sup>5</sup>*Department of Applied Physics, Graduate School of Engineering, Tohoku University, Aoba-yama 6-6-05, Sendai 980-8579, Japan*

X ray magnetic circular dichroism (XMCD) is utilised to explore the temperature dependence of the interface moments in Co<sub>2</sub>MnSi (CMS) thin films capped with aluminium. By increasing the thickness of the capping layer we demonstrate enhanced interface sensitivity of the measurements and the existence of a thin Mn oxide layer at the CMS/Al interface even when a thick capping layer is used. We show that for well ordered L2<sub>1</sub> CMS films there is no significant variation in either the Co or Mn interface moments as a function of temperature. However, a dramatic reduction in the interface moments at low temperature is observed in a disordered CMS film that is likely to be caused by increased Mn-Mn antiferromagnetic coupling. It is suggested that for ordered L2<sub>1</sub> CMS films the temperature dependence of the tunneling magnetoresistance is not related to changes in the interface moments. However, the existence of residual Mn oxide at the CMS/barrier interface could be a contributing factor.

PACS number(s): 75.70.-i, 78.70.-Dm, 72.25.-b, 75.47.-m

The possibility of exploiting the predicted half-metallic properties of full Heusler alloys of the form  $\text{Co}_2\text{MnX}$  (where  $X = \text{Si, Ge, Al}$  etc.) to obtain highly spin-polarized devices, has stimulated much interest in these materials<sup>1,2</sup>. However, only fairly recently evidence of large spin polarization has been observed in Heusler based structures, via the tunneling magnetoresistance (TMR) effect<sup>3,4</sup>. Although very high TMR values of up to 570% have been recorded at low temperatures, this value drops to around 100% at room temperature (RT)<sup>5</sup>. Understanding the origin of this temperature dependence of the spin polarization is now essential to progress work towards obtaining 100% spin-polarized devices.

There are many factors that could contribute to the temperature variation of the spin-polarization including phonon and magnon assisted tunneling, and spin-scattering processes within the barrier layer and at the interface<sup>6,7</sup>. A further possibility is that the temperature dependence is related to a change in the interface magnetization. SQUID magnetometry measurements performed on these samples do not show any significant variation in the film magnetization as samples are cooled below RT<sup>4,8</sup>. However, such measurements record only the bulk moments and are largely insensitive to any changes in the interface magnetization. As it is the interface that determines the spin-transport (e.g., TMR) properties of tunnel junctions, an interface sensitive probe of the magnetization versus temperature is required. In this Letter, we demonstrate a method for obtaining interface-specific moments and use it to explore the temperature dependence of Co and Mn moments at the interface of epitaxial CMS electrodes used for magnetic tunnel junctions. By measuring epitaxial samples we avoid complications to the temperature dependence of the magnetization caused by intergranular exchange interactions in polycrystalline CMS films<sup>9</sup>.

X ray magnetic circular dichroism (XMCD) measured using the total electron yield (TEY) technique has been shown previously to be a good method for obtaining element-specific magnetic moments<sup>10</sup>. In a  $3d$  transition metal in vacuum the TEY signal is generated by secondary electrons following x ray photon absorption. The region probed with this detection method is limited to a depth of  $\sim 2\text{-}3$  nm from the surface due to inelastic scattering of the secondary electrons. The addition of a capping layer to the metal surface will further restrict the probing depth in a way that depends on the thickness and material of the layer.

The surface sensitivity of the technique can be improved by aligning the sample at grazing incidence to the x-ray beam. However this leads to undesirable electron yield artefacts which restrict the measurement of reliable XMCD spectra. As an alternative, in this study we use the thickness of the capping layer as a method for enhancing the interface sensitivity.

Although the XMCD measured in this way does not suffer from electron yield artefacts, it is likely that the thicker capping layer could lead to saturation effects in the film<sup>11</sup>. Therefore the absolute values of moments calculated from the XMCD are not likely to be comparable to bulk measurements. We thus restrict this study to relative rather than absolute measurements of the interface moments as a function of temperature.

The CMS electrodes examined were epitaxial thin films with (100) orientation on Cr-buffered MgO(100) substrates, prepared at ambient temperature using inductively coupled plasma (ICP) assisted magnetron sputtering. A composition-adjusted sputtering target (43.7% Co, 27.95% Mn, and 28.35% Si) was used to achieve stoichiometric CMS film composition. The CMS layer was annealed *in-situ* at 450°C before depositing an Al capping layer. This structure is identical to that used in the bottom electrode of the full magnetic tunnel junctions. The overall sample structure was of the form MgO(100) / Cr(100) [40 nm] Co<sub>2</sub>MnSi(100) [30 nm] / Al [*x* nm], where *x* was varied from 2 to 4 nm. Bulk x ray diffraction measurements (not shown) were used to confirm the samples had the desired L2<sub>1</sub> superlattice structure after annealing. In addition a pair of samples was prepared in the as-deposited state and after annealing, and capped with an Al layer of 1.3 nm in both cases.

X ray absorption spectroscopy (XAS) and XMCD measurements were performed at the Advanced Light Source on beamline 6.3.1, providing 65% circularly polarized soft x rays. The x ray absorption spectra were collected using TEY detection with the sample surface inclined at 30° to the x ray beam. XMCD was collected at fixed photon helicity by reversing the magnetic field of 500 Oe along the x ray beam, for each photon energy point in the scan.

The XAS measured across the Mn *L*<sub>2,3</sub> absorption edges as a function of increasing Al capping layer thickness is shown in Fig. 1(a). It can be seen that a weak multiplet structure appears in the XAS when the Al thickness is ≥3.5 nm. This multiplet structure is characteristic for localized Mn ions in a *d*<sup>5</sup> ground state, such as in MnO, and has been observed previously in CMS thin films<sup>9,12</sup>. In addition there is a slight shift in the position of the XAS *L*<sub>3</sub> peak for thicker capping layers, although the position of the XMCD *L*<sub>3</sub> peak (not shown) does not shift. MnO is paramagnetic at RT and will therefore not contribute to the XMCD, but does yield an XAS spectrum shifted at higher energy compared to the corresponding metal. Thus the observed changes indicate the formation of a thin layer of MnO at the CMS/Al interface. It is likely that this interface layer exists for all the samples but its contribution to the XAS is only significant when the probing region is limited to the CMS/Al interface.

The relative proportion of Mn oxide within the region probed in the measurement can be assessed by measuring the XAS branching ratio,  $B = I(L_3)/[I(L_2)+I(L_3)]$ , where  $I(L_2)$  and  $I(L_3)$  are the integrated intensities over the  $L_2$  and  $L_3$  peaks, respectively<sup>13</sup>. For an oxide film the branching ratio is larger than in the native metal due to increased  $3d$  spin-orbit interaction and  $3d$  localization<sup>14</sup>. The variation of the branching ratio with Al thickness is shown in the inset to Fig. 1(a) and indicates an increase in the contribution of MnO to the XAS spectrum as the Al thickness is increased. This confirms the expected enhanced interface sensitivity as a function of capping thickness and implies that a thin layer of MnO persists at the interface even when a relatively thick capping layer is used.

The corresponding spectra measured across the Co  $L_{2,3}$  absorption edges are shown in Fig. 1(b). In this case the spectra are almost identical for all Al thicknesses used and the branching ratio remains virtually constant. Hence there is no indication of a corresponding interface layer of Co oxide in the samples. This result is not unexpected as Mn has a greater affinity for oxygen than Co and so is more likely to form an interface oxide.

The element-specific moments were derived from the XMCD for the film with the thickest capping layer in order to obtain the greatest interface sensitivity. In the following discussion we refer to these measurements as interface-specific moments. In addition, we measured CMS films with a 1.3 nm capping layer in the as-prepared (disordered) state and after annealing to obtain the  $L2_1$  superlattice structure. For these latter samples the probing depth will be greater from the interface and we thus describe these measurements as near-interface moments. In all cases the relative moment was obtained by applying sum rule analysis to the XMCD and XAS spectra using procedures discussed extensively elsewhere<sup>15</sup>. The moments thus obtained for the ordered CMS films were normalised to 1 at RT. The moments obtained from the disordered CMS film were scaled relative to the RT moment measured in the corresponding ordered sample.

The relative Co and Mn moments determined as a function of temperature, are shown in Fig. 2. It can be seen that, within the error bar, there is no significant trend with temperature for either the interface-specific or near-interface moments in the  $L2_1$  ordered films. Thus we can find no evidence to suggest that the interface magnetization contributes to the temperature dependence of the TMR in these films. However, it is interesting to note that a very different behavior is observed for the disordered CMS film (Fig. 2). In this case a dramatic decrease in both the Co and Mn moments can be seen as the temperature is reduced. This behavior is opposite to that observed for disordered CMS films by Schmalhorst *et al.*<sup>9</sup>. However, in the

previous work the temperature dependent magnetization is dominated by the onset of superparamagnetism in the polycrystalline films<sup>9</sup>. For an epitaxial CMS film in the disordered state, there is an increased probability of Mn-Mn nearest neighbour interactions with a corresponding antiferromagnetic coupling. Hybridization between Co and Mn atoms then results in a reduced moment on both sites, as has been seen previously<sup>4,9</sup>. As the sample is cooled below its effective Néel temperature the antiferromagnetic coupling will dominate over ferromagnetic coupling leading to an overall reduction in average moments. Thus the existence of site disorder in the CMS film is evidenced by a reduction in the interface moments as the temperature is decreased. The absence of such a variation in the interface-specific moments in the L2<sub>1</sub> CMS film suggests that a high degree of site ordering must extend into the interface region.

In summary, we have utilized the thickness of the capping layer in CMS/Al structure to probe the temperature dependence of the interface-specific moments. No evidence of a temperature variation is found for the ordered CMS films. These results thus rule out the possibility that the temperature dependence of the TMR could be related to variations in the interface magnetization, provided that the CMS interface has the desired L2<sub>1</sub> structure. Recent theoretical work points instead to the existence of so-called non-quasiparticle states and their role in temperature dependent transport for these structures<sup>8</sup>. However, in this study we found evidence of residual MnO at the CMS/Al interface, even when a thick capping layer was used. Thus the contribution of spin-flip scattering in this MnO layer cannot be completely neglected.

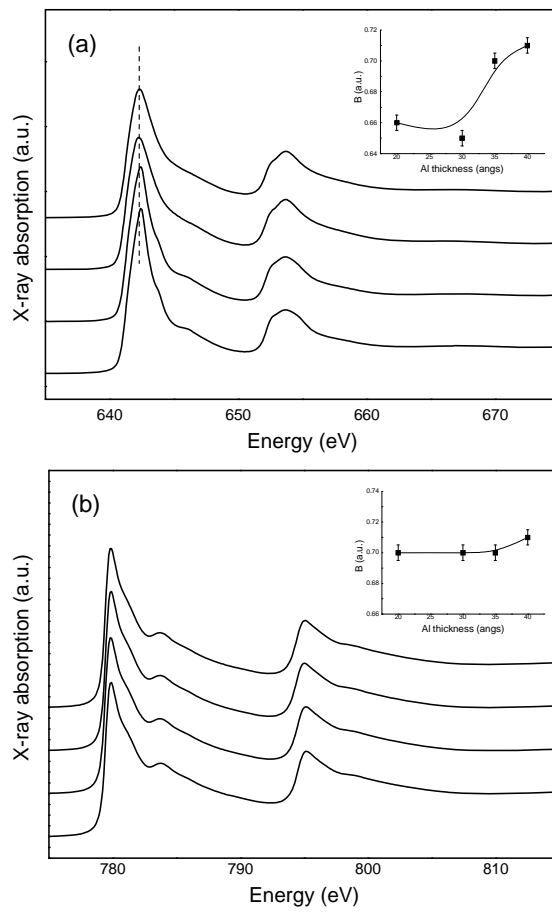
### **Acknowledgements**

The Advanced Light Source is supported by the Director, Office of Science, Office of Basic Energy Sciences, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231. A part of this work was supported by the Grant Program of the New Energy and Industrial Development Organization (NEDO) and by a Research Fellowship for Young Scientists from the Japan Society for the Promotion of Science (JSPS).

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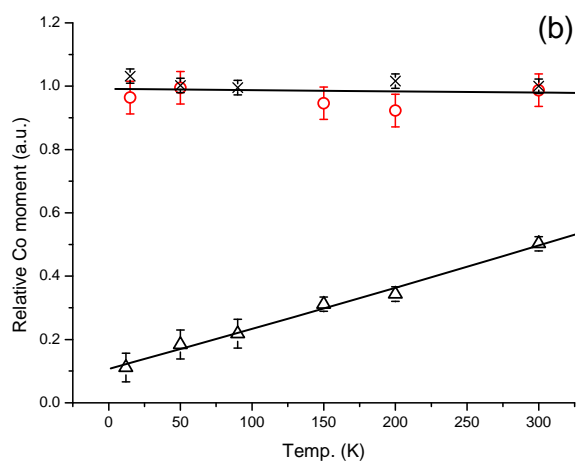
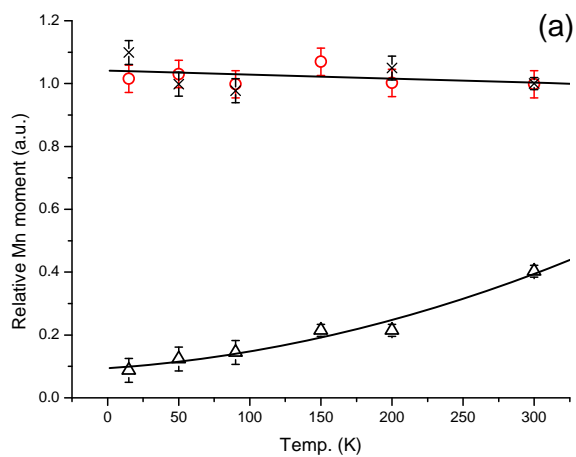
**Fig. 1: X ray absorption spectra measured across (a) the Mn and (b) the Co  $L_{2,3}$  absorption edges for CMS thin films capped with aluminium. The thickness of the Al capping layer increases from top to bottom as 2, 3, 3.5, and 4 nm. The insets show the XAS branching ratio as a function of Al thickness.**

**Fig. 2: (Color online) Relative moments derived from XMCD sum rule analysis as a function of temperature for (a) Mn and (b) Co sites. The open circles (red) are the values obtained for the 4 nm Al capping layer film (interface-specific moments) and the crosses are values obtained from the 1.3 nm capped film (near-interface moments). Also shown are the moments obtained from the disordered CMS film capped with 1.3 nm Al (open triangles).**



*Fig. 1, Telling et al*





*Fig. 2, Telling et al*