Temperature dependence of the interface moments in Co_2MnSi thin films

N. D. Telling,[1,a)] P. S. Keatley,[2] L. R. Shelford,[2] E. Arenholz,[3] G. van der Laan,[1,4] R. J. Hicken,[2] Y. Sakuraba,[6] S. Tsunegi,[5] M. Oogane,[5] Y. Ando,[5] K. Takanashi,[6] and T. Miyazaki[7]

[1]*Magnetic Spectroscopy Group, STFC Daresbury Laboratory, Warrington WA4 4AD, United Kingdom*
[2]*School of Physics, University of Exeter, Stocker Road, Exeter EX4 4QL, United Kingdom*
[3]*Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*
[4]*Diamond Light Source, Chilton, Didcot OX11 0DE, United Kingdom*
[5]*Department of Applied Physics, Graduate School of Engineering, Tohoku University, Aoba-yama 6-6-05, Sendai 980-8579, Japan*
[6]*Institute for Materials Research, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai 980-8577, Japan*
[7]*New Industry Creation Hatchery Center, Tohoku University, Aoba-yama 6-6-10, Sendai 980-8579, Japan*

X-ray magnetic circular dichroism (XMCD) is utilized to explore the temperature dependence of the interface moments in Co_2MnSi (CMS) thin films capped with aluminum. By increasing the thickness of the capping layer, we demonstrate enhanced interface sensitivity of the measurements. $L2_1$-ordered CMS films show no significant temperature dependence of either the Co or Mn interface moments. However, disordered CMS films show a decreased moment at low temperature possibly caused by increased Mn–Mn antiferromagnetic coupling. It is suggested that for ordered $L2_1$ CMS films the temperature dependence of the tunneling magnetoresistance is not related to changes in the interface moments. © *2008 American Institute of Physics*.
[DOI: 10.1063/1.2927482]

The possibility of exploiting the predicted half-metallic properties of full Heusler alloys of the form Co_2MnX (where X=Si, Ge, Al etc.) to obtain highly spin-polarized devices, has stimulated much interest in these materials.[1,2] However, only fairly recently, evidence of large spin polarization has been observed in Heusler based structures, via the tunneling magnetoresistance (TMR) effect.[3,4] 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).[5] Understanding the origin of this temperature dependence of the spin polarization is now essential to progress work toward 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.[6,7] A further possibility is that the temperature dependence is related to a change in the interface magnetization. Superconducting quantum interference device magnetometry measurements performed on these samples do not show any significant variation in the film magnetization as samples are cooled below RT.[4,8] 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 Co_2MnSi (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.[9]

X-ray magnetic circular dichroism (XMCD) measured using the total electron yield (TEY) technique has been previously shown to be a unique method for obtaining element-specific magnetic moments.[10] In a 3*d* 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 ~2–3 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 artifacts 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 artifacts, it is likely that the thicker capping layer could lead to saturation effects in the film.[11] 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 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 *in situ* annealed at 450 °C before depositing an Al capping layer. The overall sample structure was of the form MgO(100)/Cr(100) [40 nm]/Co_2MnSi(100) [30 nm]/Al[x nm], where x was varied from 2 to 4 nm. Following preparation of the Al cap-

a)Electronic mail: n.d.telling@dl.ac.uk.

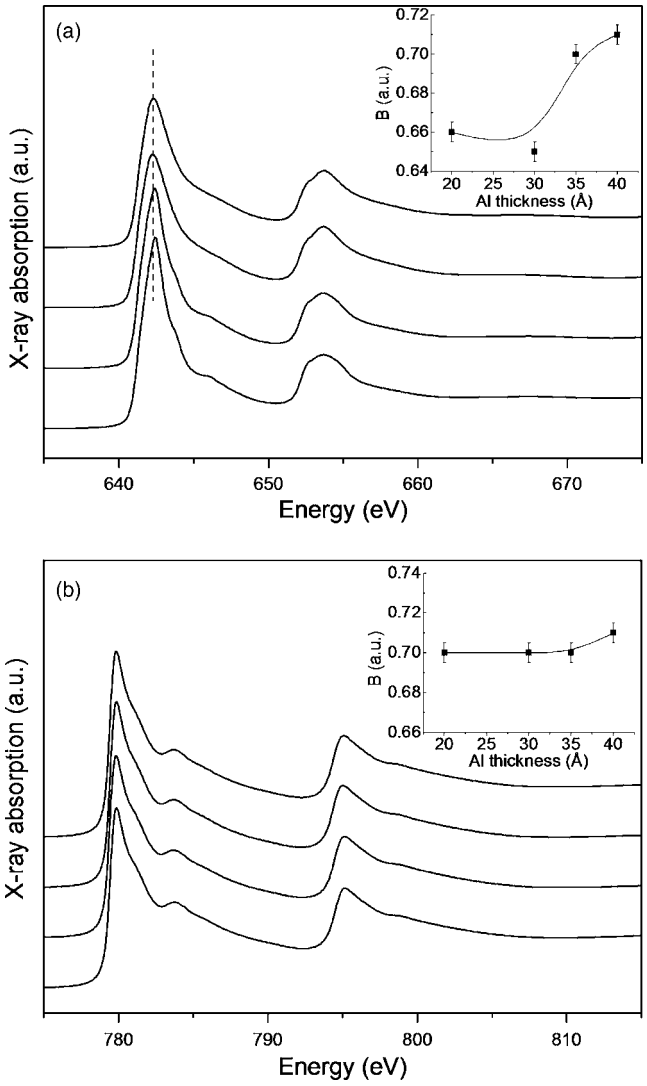

FIG. 1. X-ray absorption spectra measured across (a) the Mn and (b) the Co $L_{2,3}$ absorption edges for CMS thin films capped with aluminum. The thickness of the Al capping layer increases from top to bottom as 2, 3, 3.5, and 4 nm. The insets show the XA branching ratio as a function of Al thickness.

ping layers, the samples were allowed to naturally oxidize in air. Bulk x-ray diffraction measurements (not shown) were used to confirm the samples had the desired $L2_1$ superlattice structure after annealing. Additional samples were measured in the as-deposited state and after annealing, capped with an Al layer of 1.3 nm in both cases.

X-ray absorption (XA) spectroscopy and XMCD measurements were performed at the Advanced Light Source on beamline 6.3.1, providing 65% circularly polarized soft x-rays. The XA 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 in a scan.

The XA spectra measured across the Mn $L_{2,3}$ absorption edges as a function of increasing Al capping layer thickness are shown in Fig. 1(a). A weak multiplet structure appears in the XA spectra when the Al thickness is $\geqslant$3.5 nm. This multiplet structure is characteristic for localized Mn ions in a d^5 ground state, such as in MnO, and has been previously observed in CMS thin films.[9,12] In addition, there is a slight shift in the position of the Mn L_3 peak for thicker capping layers, although the position of the XMCD L_3 peak (not shown) does not shift. MnO is paramagnetic at RT and will therefore not contribute to the XMCD signal, but does yield an XA 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 in these naturally oxidized samples. It is likely that this interface layer exists for all capping layer thicknesses but its contribution to the XA spectra is only significant when the probed region is limited to the CMS/Al interface. For tunnel junction samples, we have previously shown that a thin layer of Mg between the CMS layer and plasma oxidized barrier layer is effective at suppressing the formation of MnO at the interface.[12]

The relative proportion of Mn oxide within the region probed in the measurement can be assessed by measuring the XA 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.[13] For an oxide film, the branching ratio is larger than in the native metal due increased $3d$ spin-orbit interaction and $3d$ localization.[14] 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 XA spectrum as the Al thickness is increased. This confirms the expected enhanced interface sensitivity as a function of capping thickness. 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 XA spectra using procedures extensively discussed elsewhere.[15] The moments thus obtained for the ordered CMS films were normalized 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.*[9] However, in the previous work the tem-

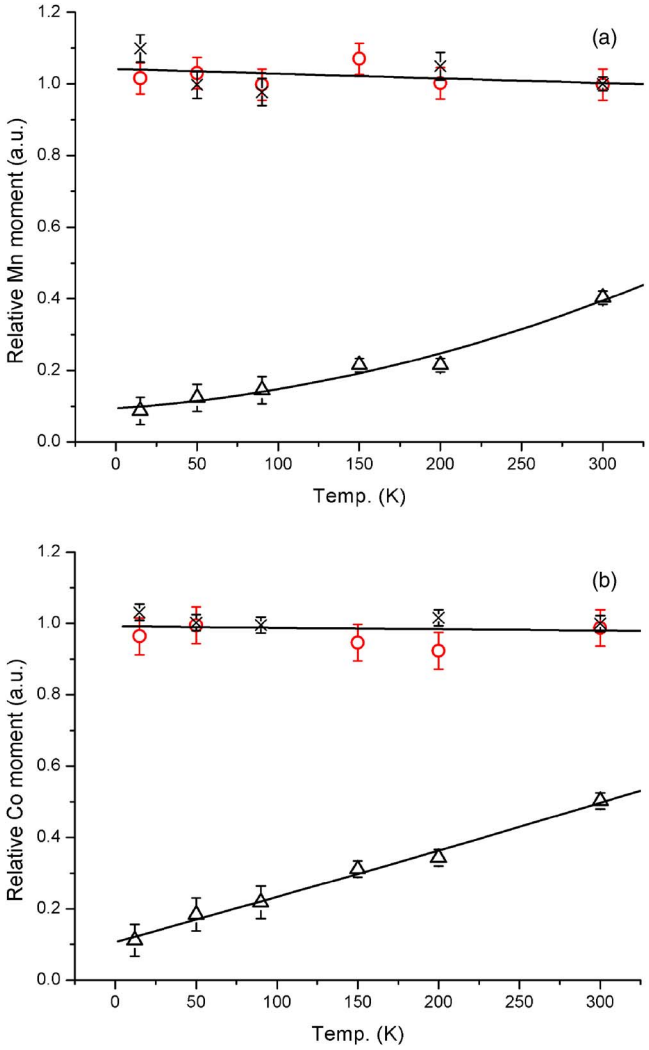

FIG. 2. (Color online) Relative moments derived from XMCD sum rule analysis as a function of temperature for (a) Mn and (b) Co sites. The open circles (red) are the values obtained for the 4 nm Al capping layer film (interface-specific moments) and the crosses are values obtained from the 1.3 nm capped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

perature dependent magnetization is dominated by the onset of superparamagnetism in the polycrystalline films.[9] For an epitaxial CMS film in the disordered state, there is an increased probability of Mn–Mn nearest neighbor interactions with a corresponding antiferromagnetic coupling. Hybridization between Co and Mn atoms then results in a reduced moment on both sites, as has been previously seen.[4,9] 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 $L2_1$ 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 structures 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 $L2_1$ structure. Recent theoretical work points instead to the existence of so-called nonquasiparticle states and their role in temperature dependent transport for these structures.[8]

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).

[1] H. C. Kandpal, G. H. Fecher, and C. Felser, J. Phys. D **40**, 1507 (2007).
[2] I. Galanakis, P. H. Dederichs, and N. Papanikolaou, Phys. Rev. B **66**, 174429 (2002).
[3] Y. Sakuraba, J. Nakata, M. Oogane, H. Kubota, Y. Ando, A. Sakuma, and T. Miyazaki, Jpn. J. Appl. Phys., Part 2 **44**, L1100 (2005).
[4] M. Oogane, Y. Sakuraba, J. Nakata, H. Kubota, Y. Ando, A. Sakuma, and T. Miyazaki, J. Phys. D **39**, 834 (2006).
[5] Y. Sakuraba, M. Hattori, M. Oogane, Y. Ando, H. Kato, A. Sakuma, H. Kubota and T. Miyazaki, Appl. Phys. Lett. **88**, 192508 (2006).
[6] C. Lü, M. W. Wu, and X. F. Han, Phys. Lett. A **319**, 205 (2003).
[7] X. Kou, J. Schmalhorst, A. Thomas, and G. Reiss, Appl. Phys. Lett. **88**, 212115 (2006).
[8] L. Choincel, Y. Sakuraba, E. Arrigoni, M. I. Katsnelson, M. Oogane, Y. Ando, T. Miyazaki, E. Burzo, and A. I. Lichtenstein, Phys. Rev. Lett. **100**, 086402 (2008).
[9] J. Schmalhorst, A. Thomas, S. Kämmerer, O. Schebaum, D. Ebke, M. D. Sacher, G. Reiss, A. Hütten, A. Turchanin, A. Gölzhäuser, and E. Arenholz, Phys. Rev. B **75**, 014403 (2007).
[10] For a general review of XMCD, see, e.g., J. Stöhr, J. Magn. Magn. Mater. **200**, 470 (1999).
[11] R. Nakajima, J. Stöhr, and Y. U. Idzerda, Phys. Rev. B **59**, 6421 (1999).
[12] N. D. Telling, P. S. Keatley, G. van der Laan, R. J. Hicken, E. Arenholz, Y. Sakuraba, M. Oogane, Y. Ando, and T. Miyazaki, Phys. Rev. B **74**, 224439 (2006).
[13] N. D. Telling, G. van der Laan, S. Ladak, and R. J. Hicken, Appl. Phys. Lett. **85**, 3803 (2004).
[14] B. T. Thole and G. van der Laan, Phys. Rev. B **38**, 3158 (1988).
[15] C. T. Chen, Y. U. Idzerda, H.-J. Lin, N. V. Smith, G. Meigs, E. Chaban, G. H. Ho, E. Pellegrin, and F. Sette, Phys. Rev. Lett. **75**, 152 (1995); B. T. Thole, P. Carra, F. Sette, and G. van der Laan, *ibid.* **68**, 1943 (1992).