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Element-specific magnetic properties of Co₂MnSi thin films

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Co₂MnSi thin films were grown on Al₂O₃ (*a* plane) and GaAs (001) substrates and on thin silicon nitride windows using pulsed laser deposition. Angle-dependent magneto-optic Kerr effect measurements reveal both a uniaxial and a fourfold magnetocrystalline anisotropy for films grown on GaAs (001). X-ray magnetic circular dichroism spectra were measured at the *L*_{2,3} edges of the thin films as a function of aluminum cap layer thickness, and transmission mode *L*_{2,3} x-ray absorption through a 1000-Å Co₂MnSi film grown on a silicon nitride membrane were measured, indicating that deviations from metalliclike spectra are likely due oxidation or contamination. Element-specific moments for Co and Mn were calculated from the X-ray magnetic circular dichroism data of a nonoxidized film. © 2005 American Institute of Physics. [DOI: 10.1063/1.1847391]

INTRODUCTION

The realization and optimization of spintronic elements such as spin valves depend on the development of highly spin-polarized materials. Half-metallic materials, i.e., those that are fully spin polarized in all bands, have been predicted through band-structure calculations, the first systems being the half-Heusler alloys NiMnSb and PtMnSb.¹ Since this discovery, interest in the Heusler alloys has regenerated, and other Heusler alloys have been predicted to be half-metallic, including Co₂MnSi and Co₂MnGe.² Although both of these systems have been predicted to be half-metallic, previous studies have reported relatively low spin polarizations, especially compared to the highly spin-polarized oxide systems CrO₂ and La_{0.7}Sr_{0.3}MnO₃.^{3–5} These disappointing results have been attributed to many factors, a common one being antisite disorder, i.e., a defect in which a magnetic atom from one sublattice in the full-Heusler *L*₂₁ structure is replaced by one from another sublattice. The moments of the displaced atoms are usually antialigned with the surrounding local moments, and therefore reduce the total magnetic moment per formula unit of the system and strongly modify the antiperiodic electron spin-polarization.^{6–8}

The magnetic, structural, and electrical properties of bulk^{9–11} and thin film^{12–15} Co₂MnSi have been extensively studied. However, the only known study of the element-specific magnetic properties of this alloy was carried out using polarized neutron reflectivity (PNR).¹¹ We have therefore employed soft x-ray magnetic circular dichroism (XMCD)

and x-ray absorption spectroscopy (XAS) to elucidate the element-specific magnetic and electronic structures of Co and Mn in Co₂MnSi.

In devices that exploit spin-polarized thin films to accomplish spin injection, the interfaces between adjacent thin-film layers in the heterostructure play a significant role. It is important that the films are of ideal crystalline structure and stoichiometry, and that deviations from their ideal structures, through disorder, strain, or oxidation/contamination, do not occur at the interface. We have studied the effects of capping films with aluminum in order to determine the effects of oxidation on the XAS and XMCD spectra on pulsed laser deposited (PLD) Co₂MnSi thin films grown on Al₂O₃ (*a* plane) and GaAs(001) substrates. In addition, polycrystalline thin films were grown on a thin (100 nm) silicon nitride window in order to measure XAS in transmission mode. Surface oxidation effects are minimized in this case since electron escape depths do not play a role in the measurement. The transmission XAS spectra were therefore used as an uncontaminated standard for comparison with the surface sensitive total electron yield (TEY) measurements.

EXPERIMENT

Thin films of Co₂MnSi were grown by ablating a single-crystal Co₂MnSi target using a KrF ($\lambda=248$ nm) excimer laser. The target to substrate distance was 35 mm and the laser repetition frequency was 10 Hz. The laser fluence (~ 4 J/cm²) was selected by positioning a focusing lens to adjust the spot size. The substrate temperature was maintained at 500 °C and the base pressure at this temperature was less than 2×10^{-8} Torr. The deposition rates were moni-

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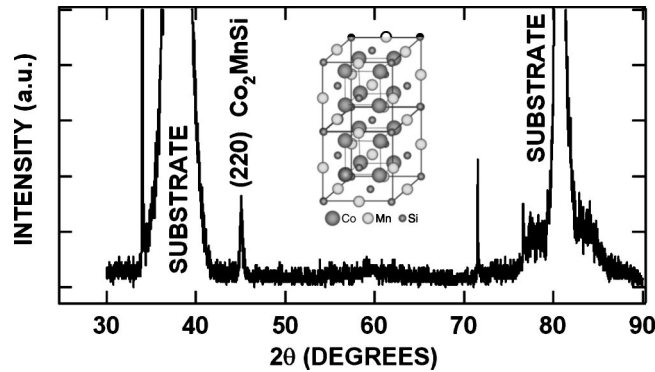


FIG. 1. XRD pattern of Co_2MnSi grown on an Al_2O_3 a -plane substrate; (inset) the $L2_1$ structure of Co_2MnSi .

tored with a quartz-crystal monitor and were calibrated using Rutherford backscattering (RBS). The film thicknesses were approximately 1000 Å (with an error of $\pm 10\%$). The films were grown on Al_2O_3 (a plane) and GaAs (001) substrates for XMCD and XAS in TEY mode, and on a silicon nitride window with a thickness of 100 nm for the XAS transmission measurements.

The soft XMCD data were acquired at the MSU X-ray Material Characterization Facility¹⁶ located at the National Synchrotron Light Source (beamline U4B) at Brookhaven National Laboratory. The U4B monochromator is of the “double dragon” design and is capable of producing highly circularly polarized (in excess of 90%) x rays in an energy range that spans 20–1350 eV.¹⁷ The polarization was set to 75% circular and $\sim 100\%$ linear for the XMCD and XAS transmission measurements, respectively. The spectra measured in TEY mode were normalized to the incident flux, and those measured in transmission mode were normalized to the incident flux and to the flux through a clean (no film) silicon nitride window of the same dimensions as that used for the thin-film samples. All the XMCD spectra were corrected for incomplete circular polarization and incident angle.

X-ray diffraction (XRD) measurements were made using a GBC DiffTech mini materials analyzer (MMA) x-ray diffractometer, and homemade magneto-optic Kerr effect apparatus was employed to acquire angle-dependent magnetometry data.

RESULTS AND DISCUSSION

The XRD measurements using $\text{Cu } K\alpha$ radiation (Fig. 1) indicated that the films grown on Al_2O_3 and GaAs were highly crystalline and had a (220) preferred orientation, consistent with the full-Heusler $L2_1$ crystalline structure (Fig. 1, inset). The peaks at approximately 34° and 71° are residual $K\beta$ reflections from the substrate. Similar results were observed for the films grown on GaAs (001). No XRD measurements were performed on the films grown on silicon nitride windows due to the delicate nature of the samples. Angle-dependent magneto-optic Kerr effect measurements were carried out to determine anisotropy constants. Magneto-optic Kerr effect (MOKE) loops as a function of in-plane orientation are shown in Fig. 2(a), demonstrating that the $[-110]$ direction is the magnetically easy axis. The

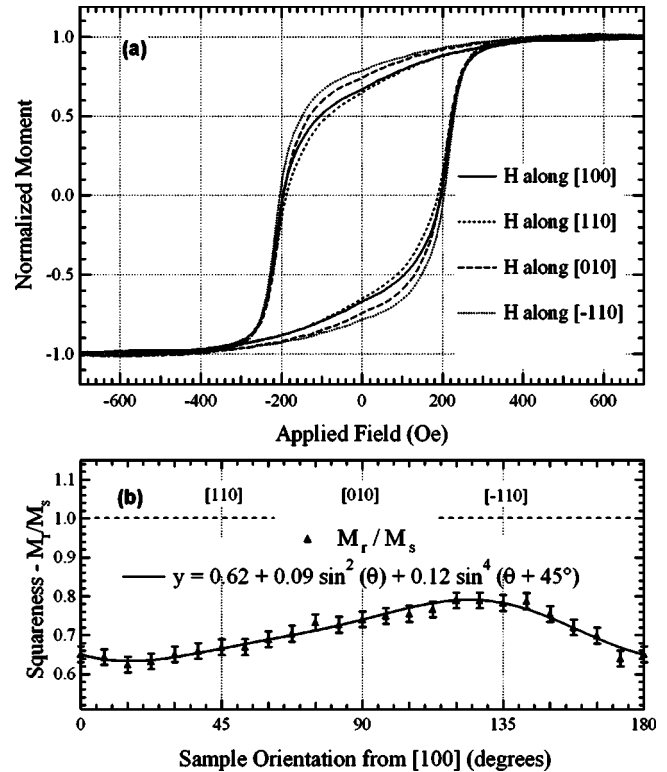


FIG. 2. (a) MOKE angle-dependent hysteresis loops and (b) the squareness plotted as a function of angle for Co_2MnSi grown on GaAs (001).

squareness (M_r/M_s) of the hysteresis loops are plotted as a function of angle in Fig. 2(b) for Co_2MnSi grown on GaAs (001). This angle dependence is consistent with a uniaxial and a fourfold anisotropy similar to that observed for Co_2MnGe films grown on GaAs (001) using molecular-beam epitaxy (MBE).^{18,19}

Co_2MnSi films (~ 100 nm) were grown on a -plane Al_2O_3 substrates and on silicon nitride windows in order to determine the degree of oxidation of the thin-film surface as a function of aluminum cap layer thickness. Multiplet structures were observed in the Mn XAS of the uncapped and 20-Å aluminum-capped samples, whereas these structures have been suppressed in the films capped with 40 Å of aluminum and in the film grown on a silicon nitride window. In transmission measurements, saturation effects are eliminated and the effects of short electron escape depths are not relevant. The transmission data are therefore sensitive to the entire film (not just the first 100 Å) and the XAS spectra looked very metallic, i.e., no multiplet features were observed, and were identical to the spectra of the 40-Å Al-capped sample. We therefore concluded that a 40-Å aluminum cap sufficiently protected the surface from oxidation and that the XMCD measured in TEY mode sampled a non-oxidized volume in this sample. We focus on the 40-Å Al-capped sample in the following discussion.

Mn and Co $L_{2,3}$ XMCD spectra of the sample capped with 40 Å of aluminum are shown in Fig. 3 and 4, respectively. These spectra are similar to those measured for Co_2MnGe and other Mn-based intermetallic alloys.^{20,21} The XMCD sum rules were used to determine the spin-orbit moment ratio for each atomic species, which is independent of

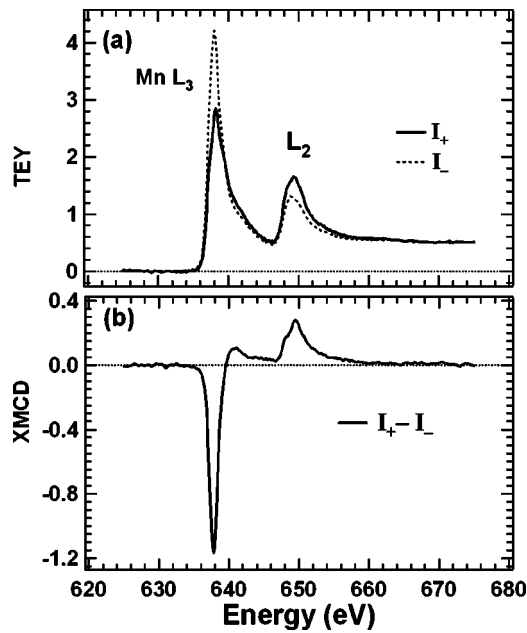


FIG. 3. (a) Polarization-dependent Mn $L_{2,3}$ TEY and (b) XMCD spectra for $\text{Co}_2\text{MnSi}/\text{Al}_2\text{O}_3$ capped with 40-Å Al.

the $3d$ electron occupation number.²² The ratio ($m_{\text{orb}}/m_{\text{spin}}$) = 0.050 and 0.022 for Co and Mn, respectively. Co and Mn $L_{2,3}$ XMCD results on Co_2MnGe (Ref. 21) are very similar to those reported here on Co_2MnSi . In particular, we observe the same doublet feature in the Mn L_2 XMCD spectrum. Although absolute moments were not estimated in the study

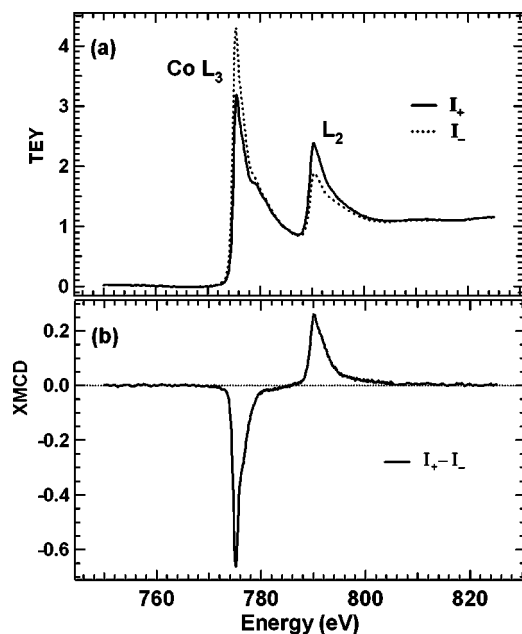


FIG. 4. (a) Polarization-dependent Co $L_{2,3}$ TEY and (b) XMCD spectra for $\text{Co}_2\text{MnSi}/\text{Al}_2\text{O}_3$ capped with 40-Å Al.

on Co_2MnGe , the spin-orbit ratios were calculated to be ($m_{\text{orb}}/m_{\text{spin}}$) = 0.10 and 0.04 for Co and Mn, respectively. In comparison, the values that were extracted for Co_2MnSi were roughly half those of Co_2MnGe .²¹

In summary, we have grown Co_2MnSi thin films and determined that multiplet structure observed in the XAS spectra is likely a result of surface oxidation, although the shape of the XMCD spectrum remains essentially unaffected. A uniaxial and a fourfold magnetocrystalline anisotropy was observed in the films grown on GaAs (001). We have estimated the spin/orbit moment ratios of Co and Mn through the application of the XMCD sum rules.

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