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Moti R. Paudel
Southern Illinois University Carbondale

Christopher S. Wolfe
Southern Illinois University Carbondale

Heather M.A. Patton
Southern Illinois University Carbondale

Jeff Simonson
Southern Illinois University Carbondale

Igor Dubenko
Southern Illinois University Carbondale

See next page for additional authors

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Authors
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Magnetic anisotropy of $Co_2MnSn_{1-x}Sb_x$ thin films grown on GaAs (001)

Moti R. Paudel,[1] Christopher S. Wolfe,[1] Heather M. A. Patton,[1] Jeff Simonson,[1]
Igor Dubenko,[1] Naushad Ali,[1] and Shane Stadler[2,a)]
[1]*Department of Physics, Southern Illinois University, Carbondale, Illinois 62901, USA*
[2]*Department of Physics and Astronomy, Louisiana State University, 202 Nicholson Hall, Baton Rouge, Louisiana 70803-4001, USA*

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Heusler alloy $Co_2MnSn_{1-x}Sb_x$ (x=0.0, 0.5, and 1.0) thin films were grown on GaAs (001) substrates using pulsed laser deposition techniques. Growth parameters have been determined that result in highly magnetically anisotropic, crystalline, and oriented (001) films. The angular dependences, relative to the GaAs (001) crystallographic directions, of the coercive field $H_c(\theta)$ and the remanence $M_r(\theta)$ were determined from angle dependent magneto-optic Kerr effect (MOKE) measurements. It was found that $H_c(\theta)$ revealed higher order symmetry contributions to the magnetic anisotropy than did $M_r(\theta)$. The Fourier analysis of rotational MOKE data was used to determine the symmetry contributions to the total anisotropy.
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I. INTRODUCTION

The Heusler alloys exhibit a wide range of interesting and useful properties that include magnetocaloric effects, ferromagnetic shape-memory, giant magnetoresistance, and high spin polarization. Since the theoretical prediction of the first half-metallic material,[1] the half-Heusler alloy NiMnSb, many other Heusler alloy systems have been predicted to be half-metallic including Co_2MnGe and Co_2MnSi, and much experimental work followed.[2–8] It was found that many of the physical properties of these systems, including the degree of spin polarization, are diminished by physical occurrences in the system, the most common ones being forms of disorder including, primarily, antisite disorder, which has been observed experimentally.[9,10] For instance, it was predicted that Co antisite disorder in Co_2MnSi introduced a finite contribution in the gap of the minority density of states, resulting in suboptimum spin polarization, i.e., far below the predicted values.[11] Low spin polarization values have been reported for many of these systems including Co_2MnGe and Co_2MnSi.[7,8,12]

Materials fabricated in bulk from the melt have an inevitable degree of disorder that is driven by the thermodynamics of the fabrication process. Steps can be taken to minimize the degree of disorder: For instance, the bulk fabrication parameters can be optimized or films can be grown at low temperatures. Another approach is the fabrication of substituted Heusler alloy systems as suggested by Ishida, *et al.*,[13] where they showed that the properties of the minority gap and the placement of the Fermi level within that gap depended on the composition. One such set of quaternary alloys that have been predicted to be highly spin polarized (even half-metallic) is $Co_2MnSn_{1-x}Sb_x$, which is the subject of the current study.[13] Here we report on the fabrication of $Co_2MnSn_{1-x}Sb_x$ (x=0.0, 0.5, and 1.0) thin films using pulsed laser deposition (PLD) and their structural and magnetic (including magnetocrystalline anisotropy) properties.

II. EXPERIMENTAL

Polycrystalline buttons (approximately 5 g) of $Co_2MnSn_xSb_{1-x}$ (x=0, 0.5, and 1.0) alloys were fabricated using conventional arc-melting methods in an argon atmosphere using commercially available elements with 4N purity. To enhance the homogeneity, they were wrapped in tantalum foil and annealed in vacuum ($<10^{-4}$ Torr) at 850 °C for 24 h. For the growth of film samples, the bulk alloys were cut into thick disks (for PLD targets) and were mirror polished using sand paper in ethyl alcohol. The films were grown on American flat GaAs (100) wafers. Before film growth, the substrates were first ramped to 600 °C, and held for 15 min in the ultrahigh vacuum (UHV) deposition chamber in order to remove any oxides present on the surface and to reconstruct the surface. The substrates were subsequently cooled down to 178 °C for deposition. Prior to film deposition, the targets were conditioned with the laser in order to remove any oxides formed on the surface.

Thin films were grown in a UHV PLD chamber with a base pressure less than 5×10^{-9} Torr. A KrF (λ=248 nm) excimer laser (Lambda Physik Compex 201) with a 20 ns pulse width and a 10 Hz repetition rate was employed. The target to substrate distance was 42 mm and the laser fluence was approximately 4 J/cm^2. The deposition rates were monitored with a quartz crystal monitor and were calibrated using Rutherford backscattering (RBS) data. RBS was also used to monitor the stoichiometry of both the targets and the film samples. We concentrated on the samples grown with a deposition rate of approximately 20 Å/min and thicknesses of 150 Å with an error of ±5%.

Structural phases were identified through x-ray diffraction (XRD) measurements at room temperature using a GBC MMA XRD in Bragg–Brentano geometry using Cu $K\alpha$ ra-

a)Author to whom correspondence should be addressed. Electronic mail: stadler@phys.lsu.edu.

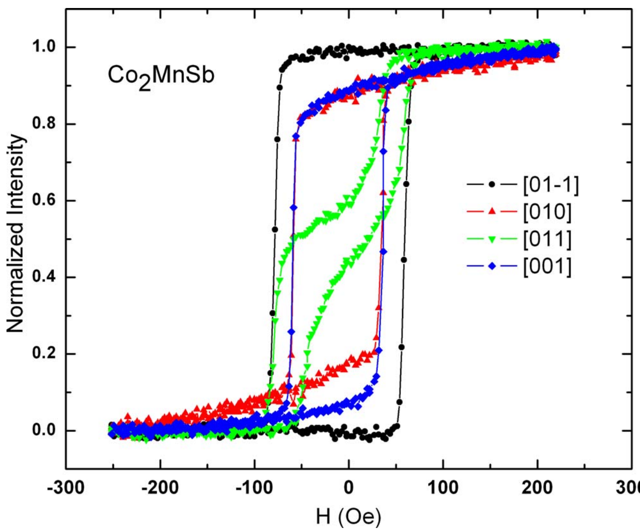

FIG. 1. (Color online) MOKE loops of $Co_2MnSb(150\ Å)/GaAs(001)$ as a function of GaAs crystallographic direction.

diation. The compositional analysis of the films was done with RBS and was further confirmed using X-ray fluorescence (XRF) spectroscopic measurements. The target compositions were confirmed with inductively coupled plasma (ICP) analysis. Longitudinal, angle-dependent, magneto-optic Kerr effect (MOKE) and rotational MOKE (ROTMOKE) measurements were made on a home-built system. The system employs a linearly polarized HeNe laser with the polarization perpendicular the scattering plane (i.e., *s*-polarization).

III. RESULTS AND DISCUSSION

Films were grown from the bulk Heusler alloy targets $Co_2MnSn_xSb_{1-x}$ (x=0, 0.5, and 1.0). Similar to the fabrication conditions reported for PLD grown Co_2MnSi (Ref. 8), it was found that a growth temperature of 178 °C (~450 K) produced films of the highest crystalline and magnetic quality, evidenced through XRD patterns and MOKE hysteresis loops (i.e., high squareness and significant anisotropy), respectively. The films for this study were 150 Å thick and, through XRD measurements (data not shown), were found to be highly (001) oriented. The compositions of the films were found to be *sp*-element deficient from RBS and XRF measurements, probably resulting from the segregation of these elements to grain boundaries in the polycrystalline target. A more congruent transfer of material from the target to the film surface could likely be accomplished by using single crystal targets.

Angle-dependent MOKE measurements were carried out on the films and they were found to be highly anisotropic. The dependence of the hysteresis loops on the crystallographic directions relative to the GaAs (001) substrates is shown in Fig. 1. Coercive fields of all of the films were approximately H_c=70 Oe along the easy axis $[01\bar{1}]$ direction. A similar dependence has been observed for Co_2MnGe Heusler alloy films of comparable thicknesses grown on GaAs (001) by molecular beam epitaxy[3] and for Co_2MnSi grown by PLD.[4,8] Starting on the easy axis, angle-dependent MOKE loops were measured at every 5° and the squareness

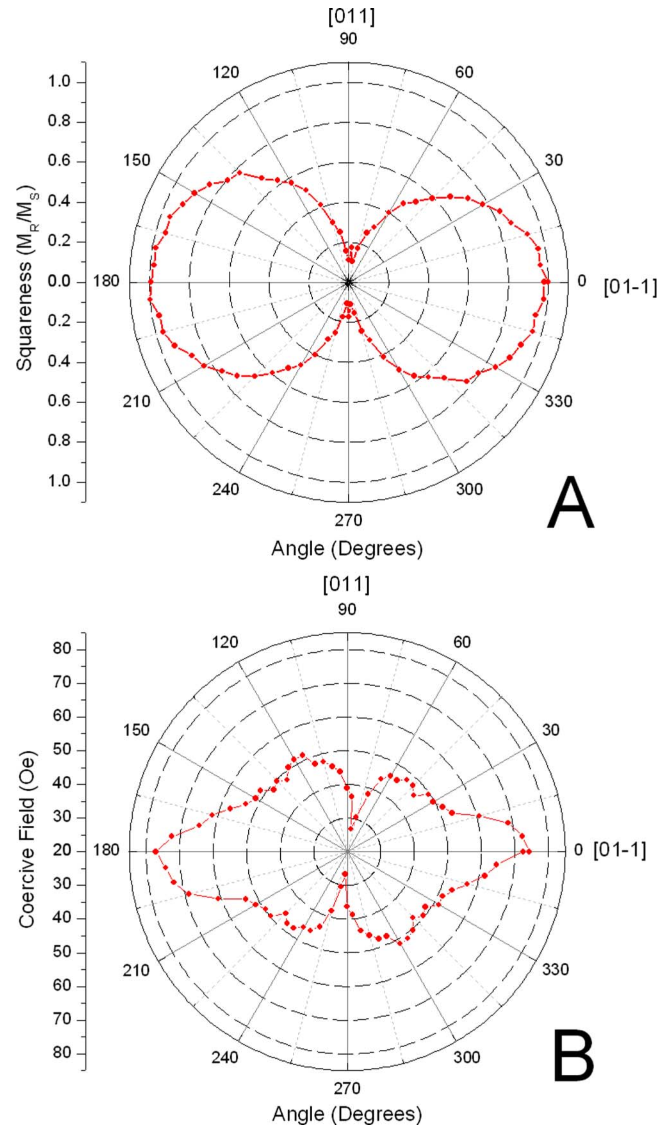

FIG. 2. (Color online) (a) Polar plot of the squareness (M_r/M_s) of $Co_2MnSb(150\ Å)/GaAs(001)$ determined from angle-dependent MOKE loops. GaAs substrate crystallographic directions are indicated. (b) Coercive field (H_c) as a function of angle for the same sample.

(M_r/M_s) of a Co_2MnSb (150 Å) film is plotted in Fig. 2(a). These systems usually show a combination of a fourfold and uniaxial anisotropy, where the fourfold component comes from the cubic ($L2_1$) symmetry of the film and the uniaxial component from the GaAs (001) substrate surface. This type of symmetry is visually apparent in the remanence (or squareness) plot of Fig. 2(a). The coercive field H_c of the same film is plotted as a function of angle in the polar plot of Fig. 2(b). In this case, additional periodic features are apparent, indicating the existence of an additional component to anisotropy. Another method was needed to verify the nature of this magnetic anisotropy.

There are many techniques that can be employed to study magnetic anisotropy; they range from ferromagnetic resonance to torque magnetometry. In this study we employed the ROTMOKE method, where the Kerr rotation is measured as a function of the angular position of an in-plane magnetic field (in this case H=400 Oe), i.e., the sample position remains constant and the field rotates (in-plane) around

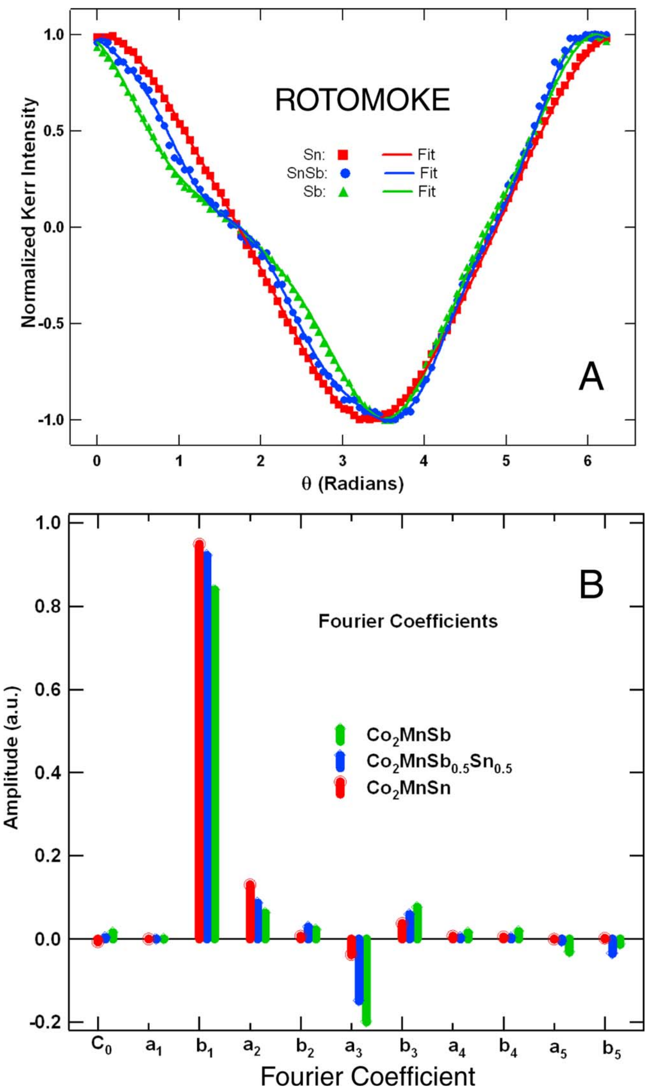

FIG. 3. (Color online) (a) ROTMOKE measurements for $Co_2MnSn_{1-x}Sb_x$ ($x=0$, 0.5, and 1) and the corresponding Fourier fits. (b) Fourier coefficients from the fits of the data in part (a).

the sample. The ROTMOKE method was introduced by Mattheis and Quednau.[14] The ROTMOKE data, starting from the hard magnetic axis, for $Co_2MnSn_{1-x}Sb_x$ ($x=0.0$, 0.5, and 1.0) are shown in Fig. 3(a). A film that possesses only an isotropic in-plane anisotropy would follow a simple cosine dependence, and a deviation from a simple cosine dependence implies the existence in-plane anisotropy. If the cosine dependence is subtracted from the data, what is left is proportional to the torque. The Fourier analysis results of the ROTMOKE data for all three samples are shown in Fig. 3(b), where C_0 is the constant term, and a_n and b_m are the coefficients of the sine and cosine terms, respectively. The b_1 coefficient is large, since we have not subtracted this dominant linear (cosine) term from the data. The a_2 and b_2 terms are attributed to the second order Kerr effects ($a_2 \sim m_L m_T$ and $b_2 \sim m_L^2 - m_T^2$). Large quadratic MOKE effects have been observed in other Heusler alloys such as Co_2FeSi.[15] The coefficients a_3 and b_3 usually reflect the threefold polar signal in the case of (111) oriented films,[16] however, there was no evidence of (111) orientation in our samples. These and other higher order coefficients can be attributed to many possible origins including strain, surface structure, and texturing.[17]

The occurrence of strain and texturing are both strong possibilities in these systems. First, there is a lattice mismatch of about 5% between the bulk alloys and the GaAs substrate, indicating a strained condition for the films as observed using XRD. Second, film growth using PLD can result in highly textured films depending on the specific growth conditions and geometry.

In conclusion, we have grown (100) oriented thin films of the quaternary Heusler alloys $Co_2MnSn_{1-x}Sb_x$ ($x=0.0$, 0.5, and 1.0) from stoichiometric targets using PLD. Angular dependent MOKE loop measurements reveal a complex in-plane magnetic anisotropy. The coercive field (H_c) as a function of angle indicates the presence of multiple-order anisotropy terms. Finally, the Fourier analysis of ROTMOKE data revealed the Fourier components contributing to the total anisotropy. All films exhibited nonlinear effects, where the Co_2MnSn ($x=0$) film deviated the least from the cosine dependence in the ROTMOKE measurements. Higher order Fourier components likely originated from strain and/or texturing during the PLD growth process.

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