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<tr>
<td>Citation</td>
<td>Journal of Applied Physics, 107(4): 043912</td>
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<tr>
<td>Issue Date</td>
<td>2010-02-15</td>
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<tr>
<td>Doc URL</td>
<td><a href="http://hdl.handle.net/2115/42761">http://hdl.handle.net/2115/42761</a></td>
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Magneto-optical investigation of epitaxial nonstoichiometric $\text{Co}_2\text{MnGe}$ thin films

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(Received 9 October 2009; accepted 16 December 2009; published online 25 February 2010)

We investigate the magneto-optical properties of a nonstoichiometric, epitaxial $\text{Co}_2\text{Mn}_{0.77}\text{Ge}_{0.42}$ (001) film grown on a MgO-buffered MgO(001) single-crystal substrate. Magneto-optical Kerr effect magnetometry indicates that the sample has strong uniaxial anisotropy, whereas the easy and hard axes of magnetization are tilted by $\sim 10^\circ$ with respect to the [110] and [110] directions, respectively. A modest quadratic Kerr effect with an amplitude of 0.4 mdeg was observed. Brillouin light scattering spectroscopy was used to find that the exchange constant $A$, spin-wave stiffness $D$, and saturation magnetization $M_s$ are 22.5 pJ/m, 413 meVÅ$^2$, and 6.43 $\mu_B$/f.u., respectively. The saturation magnetization value suggests that the Slater–Pauling rule might apply to such nonstoichiometric compositions. © 2010 American Institute of Physics. [doi:10.1063/1.3296350]

I. INTRODUCTION

$\text{Co}_2\text{MnGe}$, as many other $\text{Co}_2$-based Heusler compounds ($\text{Co}_2\text{MZ}$, where $M$ is a transition metal and $Z$ is a main group element), is attracting considerable attention due to its predicted half-metallicity.$^{1-4}$ Materials endowed with this property support a particular band structure in which the majority-spin channel is metallic, whereas the minority spin channel is semiconducting. In theory, this provides a 100% spin polarization at the Fermi level, which is attractive for spintronic applications. For $\text{Co}_2\text{MnGe}$, the Fermi level lies near the middle of the minority spin band gap.$^2$ This is advantageous for conserving half-metallicity at finite temperatures or in the presence of defects which smear the electron density near the valence or conduction bands. Furthermore, $\text{Co}_2\text{MnGe}$ boasts a high Curie temperature of 905 K,$^5$ which is a prerequisite for implementation in practical devices. It also features a small lattice mismatch of $\sim 3.6\%$ between $\text{Co}_2\text{MnGe}$ and MgO [after a 45$^\circ$ in-plane rotation within the (001) plane, see Fig. 1], compared to, for example, that of $\sim 5.1\%$ between $\text{Co}_2\text{MnSi}$ and MgO. This small mismatch is not only preferable for realizing high-performance magnetic tunnel junctions (MTJs) with potentially half-metallic Heusler-compound electrodes but also to build layered structures for spin-injection from a Heusler-compound electrode into semiconductors through a MgO barrier.

Some of the present authors recently developed fully epitaxial MgO-based MTJs wherein a Heusler-compound thin film serves as either the lower or both the lower and upper ferromagnetic electrodes.$^{6-10}$ Relatively high tunnel magnetoresistance (TMR) ratios of 376% at 4.2 K and 160% at room temperature (RT) were reported for $\text{Co}_2\text{MnGe}/\text{MgO}/\text{Co}_{50}\text{Fe}_{50}$ MTJs with a Mn- and Ge-deficient $\text{Co}_2\text{Mn}_{0.77}\text{Ge}_{0.42}$ lower electrode.$^{11,12}$ Furthermore, high TMR ratios of 1135% at 4.2 K and 236% at RT were demonstrated for $\text{Co}_2\text{MnSi}/\text{MgO}/\text{Co}_2\text{MnSi}$ MTJs with Mn-rich $\text{Co}_2\text{Mn}_{1.29}\text{Si}$ electrodes.$^{13}$

It was recently shown that the temperature dependence of the TMR ratio of a $\text{Co}_2\text{FeAl}_{0.5}\text{Si}_{0.5}/\text{MgAl}_{2}\text{O}_4/\text{CoFe}$ TMI followed a $T^{0.2}$ power law ($T$ being the temperature), due to the thermal excitation of spin waves.$^{14}$ Furthermore, for fully epitaxial $\text{Co}_2\text{MZ}/\text{MgO}$-based MTJs, including $\text{Co}_2\text{MnSi}/\text{MgO}/\text{Co}_{50}\text{Fe}_{50}$, $\text{Co}_2\text{Cr}_{0.5}\text{Fe}_{0.5}\text{Al}/\text{MgO}/\text{Co}_{50}\text{Fe}_{50}$, and $\text{Co}_2\text{MnSi}/\text{MgO}/\text{Co}_2\text{MnSi}$ (Ref. 16) junctions, the tunnel resistance for the parallel magnetization configuration were almost independent of temperature from 4.2 to 300 K, while that for the antiparallel magnetization configuration ($R_{AP}$) were strongly dependent on $T$ and decreased with increasing $T$. This strong temperature dependence of $R_{AP}$ was also...
cussed in terms of the role of thermally excited magnons for spin-flip scattering\textsuperscript{17} from the majority-spin band to minority spin gap states, including minority spin interface states, in the emitter electrode, and vice versa in the collector electrode.\textsuperscript{16,18} In this context, the knowledge of the exchange stiffness, which will largely determine the energy required to create a spin wave, is a crucial parameter that must be understood to gain insight into the construction of superior devices.

As a part of our ongoing investigation of the magneto-optical properties of Co\textsubscript{2}-based Heusler compounds by means of magneto-optical Kerr effect (MOKE) magnetometry\textsuperscript{19–23} and Brillouin light scattering (BLS) spectroscopy,\textsuperscript{19,24–26} we here report on the investigation of the Mn- and Ge-deficient, nonstoichiometric epitaxial Co\textsubscript{2}Mn\textsubscript{0.77}Ge\textsubscript{0.42} films described above. The compound of interest here, Co\textsubscript{2}MnGe, was recently investigated by BLS spectroscopy.\textsuperscript{27,28} However, in that study, Co\textsubscript{2}MnGe films were grown by rf-sputtering onto V-buffered Al\textsubscript{2}O\textsubscript{3} and the actual stoichiometry was not verified. We show the exchange constant is nearly doubled for our nonstoichiometric Co\textsubscript{2}Mn\textsubscript{x}Ge\textsubscript{1-x} (x, y < 1) thin films compared to that reported for the Co\textsubscript{2}MnGe(110) films.\textsuperscript{27,28}

II. SAMPLE PREPARATION AND CHARACTERIZATION

A. Sample preparation and structural details

The nominal studied sample layer structure was as follows: MgO(001) substrate/MgO buffer (10 nm)/Co\textsubscript{2}MnGe (50 nm)/MgO barrier (2 nm)/AlO\textsubscript{x} capping layer (1 nm). The sample was prepared in an ultrahigh vacuum chamber maintained at a base pressure of \(\sim 6\times 10^{-8}\) Pa. The Mn- and Ge-deficient Co\textsubscript{2}MnGe films were deposited at RT by radio frequency magnetron sputtering from a stoichiometric Co\textsubscript{2}MnGe target and subsequently annealed \textit{in situ} at 500 °C for 15 min. The MgO barrier was deposited by electron beam deposition at RT. During this deposition step, the pressure was \(\sim 6\times 10^{-7}\) Pa. Finally, an Al layer was deposited by direct current sputtering at RT and a native oxide was formed in an oxygen atmosphere over a period of 2 h. Further details of the sample preparation can be found in Refs. 11 and 12.

X-ray diffraction measurements show that the films grew as epitaxial single-crystal thin films, and the presence of \{111\} reflections indicate the sample is at least partially L\textsubscript{2}\textsubscript{1} ordered.\textsuperscript{29} A high-resolution transmission electron microscopy study of a cross section of similar samples also revealed that the Mn- and Ge-deficient Co\textsubscript{2}MnGe films were epitaxial and had very sharp interfaces with MgO.\textsuperscript{12} Furthermore, the \{111\} diffraction spots were also observed by electron diffraction, consistent with L\textsubscript{2}\textsubscript{1} ordering.\textsuperscript{12} Using inductively coupled plasma optical emission spectroscopy, the composition of a thin film prepared under the same conditions was determined to be Co\textsubscript{2}Mn\textsubscript{0.77}Ge\textsubscript{0.42}, with an accuracy of 2\%–3\% for each element.\textsuperscript{11,12} We assume the same stoichiometry for the sample studied here. This composition of Co\textsubscript{2}Mn\textsubscript{0.77}Ge\textsubscript{0.42} is important to understand the defects induced in off-stoichiometric Co\textsubscript{2}MnGe films and will be discussed in Sec. III B with respect to the experimentally obtained saturation magnetization for this film.

B. MOKE measurements

All MOKE and BLS measurements described below were preformed at RT.

The sample was mounted on a stage capable of rotating around an axis that is perpendicular to the sample’s surface and contained by the plane of light incidence. The measurements were carried out in the longitudinal configuration (i.e., the magnetic field is applied in the sample plane and is parallel to the plane of light incidence). The incident light (laser diode, \(\lambda = 670\) nm) was plane polarized perpendicular to the plane of light incidence (s-polarized light). The sample was oriented such that the edge of the substrate (i.e., the MgO [110] direction) is aligned with the 0° in-plane orientation. That is to say, 0° is along the Co\textsubscript{2}MnGe [110] direction (see Fig. 1). For a given in-plane orientation \(\alpha\) (with respect to the magnetic field) of the sample, the Kerr rotation is measured as the applied magnetic field is cycled between \(\pm 35\) mT. The sample is then rotated by 1°, and another Kerr rotation versus field loop is measured. This procedure is repeated until a loop has been measured for every in-plane orientation through a full rotation of the sample (360°). The field strength of 35 mT was sufficient to magnetically saturate the sample, for all orientations.

C. QMOKE measurements

These measurements were carried out using a MOKE magnetometer equipped with a quadrupole magnet, which is described elsewhere.\textsuperscript{53} Only results obtained at perpendicular incidence are shown. Briefly, for a given sample orientation, the Kerr rotation is measured in saturation (50 mT) along eight field directions, each oriented 45° away from one another. The Kerr rotation \(\phi_{K}\) is measured along the field labeled \(H_{1}\), which is oriented at 45° (in a clockwise fashion) from the plane of light incidence on the side of the reflected light beam. The subsequent fields labeled 2–8 are successively found in a clockwise fashion. This procedure is repeated every incremental rotation of 5° through a full rotation of the sample. As will be explained below and is detailed in Refs. 20, 30, and 31, the combination of these eight Kerr rotations \(\phi_{K}\) allows one to determine the contributions to the MOKE signal that are proportional to the longitudinal component of the magnetization \(M_{L}\), as well as product terms containing both the longitudinal and the transverse components of the magnetization, \(M_{T}\).

D. BLS measurements

The BLS measurements were performed in the magnetostatic surface mode geometry, i.e., the magnetic field \(H\) was applied parallel to the film surface and perpendicular to the plane of light incidence. A diode-pumped, frequency-doubled Nd:YVO\textsubscript{4} laser (\(\lambda = 532\) nm) was used as a light source. The angle of incidence \(\phi\) (defined by the direction of the incident light and the sample’s normal) and the magnetic field were changed. A more detailed description of this instrument is found in Ref. 32.
Except where specifically indicated, the BLS spectra were recorded at an angle of incidence $\varphi$ of 45°. For a given angle of incidence, the in-plane wave vector $q_\parallel$ of the detected magnons is given by

$$q_\parallel = \frac{4\pi}{\lambda} \sin(\varphi).$$

### III. RESULTS AND DISCUSSION

#### A. MOKE magnetometry

The coercive fields determined for each in-plane sample orientation are compiled in Fig. 2(a). We remind that in this figure the 0° orientation corresponds to the Co$_2$MnGe [1 1 0] orientation (see Fig. 1).

The coercive field $\mu_0H_c$ ranges between 1.15 and 1.61 mT. The angular dependence clearly displays a uniaxial profile, where the easy axis of magnetization (highest coercive field) is oriented about 7° past the [1 1 0] direction (i.e., $\alpha \sim 97°$), and the in-plane hard axis is oriented orthogonally to this direction.

For loops measured at orientations where larger coercivities were observed, for example, at angles of 75° and 97°, as shown in Figs. 3(d) and 3(e), the Kerr rotation loop was quite square, confirming the easy axis character of that orientation. Conversely, loops measured in orientations of lower coercivity showed rounding, which reflects its hard direction character. Such loops can be seen in Figs. 3(b), 3(c), 3(f), and 3(h).

More generally, the Kerr rotation measured in remanence $\phi_r$, shown in Fig. 2(b) (as well as the reduced remanence $\phi_r/\phi_{sat}$, not shown) shows the same uniaxial behavior as the coercive field shown in Fig. 2(a), with the remanence (and reduced remanence) being maximal along the [1 1 0] direction. This again indicates that the easy axis of magnetization is oriented along the [1 1 0] direction. The fine structure observed in Fig. 2(a) between 130° and 220° is also more apparent in this graph.

The origin of such uniaxial symmetry may be hard to assess. The small step between our measurements also highlights the rather complex shape of the anisotropy, a feature that is often lost when fewer measurements are performed. To explain the experimental value of the saturation magnetization of 6.43 $\mu_B$/f.u., we tentatively estimated the formula unit composition to be Co$_2$[Mn$_{0.492}$Co$_{0.508}$][Ge$_{0.527}$Mn$_{0.473}$], as will be described in Sec. III B, where the first bracket relates to the site occupancy of the Mn site of the ideal L2$_1$ structure, and the second bracket relates to the Ge site occupancy. We speculate that the observed uniaxial symmetry was probably related to the fact that (1) the Ge sites were only half-occupied by Ge atoms in the Co$_2$Mn$_{0.77}$Ge$_{0.42}$ film, and (2) a possible preferential distribution of the Ge atoms on the Ge sites is present.

When strains lower than 2% were present, for example, in Co$_2$Cr$_{0.6}$Fe$_{0.4}$Al grown on Fe- or Cr-buffered MgO (Ref. 22) and Co$_2$MnSi grown on Cr-buffered MgO$^{10}$, we previously observed a very clear fourfold anisotropy, which was indicative of the cubic magnetocrystalline anisotropy of these samples.

Studies of the magnetic properties of single-crystal (001) oriented Co$_2$MnGe thin films on GaAs(001) have revealed a combination of in-plane fourfold and uniaxial anisotropy.$^{33,34}$

![Diagram of Kerr rotation loops measured in fields of ±35 mT.](image-url)
However, in both these cases, multistep magnetization reversal was observed, which was attributed to two combined anisotropy (cubic magnetocrystalline anisotropy and surface uniaxial anisotropy) sharing the same easy axis. This is not the case here, as can be seen in Fig. 3 where magnetization reversal is seen to occur in a single step for every orientation.

As can be seen in Fig. 4 for measurements at an in-plane orientation of $173^\circ$, the measured Kerr rotation exhibits an asymmetry with respect to an inversion of the magnetic field. The symmetric and asymmetric components of the Kerr signal $\phi$ may be arithmetically extracted.

The two components are given as

$$\text{LMOKE}_{|\pm\rangle} \rightarrow \phi_{\text{sym}} = \frac{\phi(H_{\pm\text{+}}) - \phi(-H_{\pm\text{+}})}{2},$$

$$\text{QMOKE}_{|\pm\rangle} \rightarrow \phi_{\text{asym}} = \frac{\phi(H_{\pm\text{+}}) + \phi(-H_{\pm\text{+}})}{2},$$

where the arrows indicate the branches of the loop with increasing ($\uparrow$) and decreasing ($\downarrow$) field strengths. As is indicated, the component that is symmetric upon field inversion is attributed to the longitudinal component of the Kerr signal (LMOKE), and the asymmetric component is attributed to a quadratic component (QMOKE). The origin of QMOKE is not yet fully understood, but the contemporary view places its origin in second-order spin-orbit coupling. The microscopic details leading to the observation of QMOKE in a given sample are not yet firmly established, but QMOKE has been observed in a range of cobalt-based Heusler-compound thin films such as Co$_2$MnGe$_{1-x}$Si$_x$(111), Co$_2$FeSi(001), Co$_2$MnSi(001), and (among other systems) thin Fe films.

To quantify the amplitude of the QMOKE component, we have carried out the “eight field method” (see Sec. II and Refs. 20, 30, and 31 for more details). The eight measured Kerr rotations $\phi_n$ are combined according to (see Fig. 1 for geometry)

$$\phi_{\text{sat}}^{\text{QMOKE}} = \frac{\phi_8 - \phi_4}{2},$$

and yield the LMOKE signal in saturation $\phi_{\text{sat}}^{\text{LMOKE}}$ as well as two QMOKE signals $\phi_{\text{sat}}^{\text{QMOKE}, a}$ and $\phi_{\text{sat}}^{\text{QMOKE}, b}$. The measured signals are presented in Fig. 5 as a function of in-plane sample orientation. As can be seen, the LMOKE does not depend on the sample direction. The two QMOKE signals exhibit a sinusoidal behavior, and both have the same amplitude (0.4 mdeg in this case) and the MOKE signal proportional to $M_L M_T$ is vertically offset from zero. This amplitude, while clearly observed experimentally due to our sensitive experimental setup, is quite modest. For comparison, amplitudes of $\sim 2$ and $\sim 18$ mdeg were observed for Co$_2$MnSi (Ref. 23) and Co$_2$FeSi (Ref. 20) thin films, respectively.

The expected Kerr rotation for a cubic material, when taking into account effects up to the second order of the magnetization, is the real part of

$$\Phi_2 = A \cdot K M_L + B \left[ \left( \frac{K}{\eta^2} + 2 G_{44} + \frac{\Delta G}{2} \right) M_L M_T - \frac{\Delta G}{2} \cos 4 \alpha \cdot M_L M_T - \frac{\Delta G}{4} \sin 4 \alpha (M_L^2 - M_T^2) \right],$$

where $A$ and $B$ are the (complex) optical weighting factors, $K$ and $G_{ij}$ are the components of the linear and quadratic magneto-optical tensors, respectively, $\Delta G = G_{11} - G_{12} - 2G_{44}$ embodies the magneto-optical anisotropy, and $\eta$ is the complex refractive index of the material. The salient features of Eq. (7) are recognized in the data shown in Fig. 5. This suggests that the electronic properties of this sample are well described by a cubic environment, as expected for a Heusler compound. The solid lines in Fig. 5 are fits to the various components of Eq. (7).

**B. BLS spectroscopy**

BLS spectra were measured in various magnetic fields, as is shown in Fig. 6(a). The positions of the observed peaks shift to larger absolute frequencies with increasing field. This
is evidence that these peaks are of magnonic origin, as peaks arising from phonons would not be shifted by an applied magnetic field.\textsuperscript{41}

The Damon–Esbach mode (dipole-dominated magneto-static surface mode, i.e., \( q \perp M_s \)) can be discriminated from the perpendicular standing spin-wave (PSSW) mode by changing the angle of incidence. For the former (neglecting exchange), the spin-wave frequency \( \omega_{\text{DE}} \) is given in Système International units by

\[
\omega_{\text{DE}} = \gamma \mu_0 \sqrt{H(H + M_s) + \frac{M_s^2}{8\pi}(1 - e^{-2qd})},
\]

where \( \gamma=(1/g_e)\gamma_g|g \) is the gyromagnetic ratio (\( \gamma_g \) is the gyromagnetic ratio of a free electron, and \( g_e \) and \( g \) are the Landé factors for the free electron and the material, respectively), \( \mu_0 \) is the permeability of vacuum, \( M_s \) is the saturation magnetization\textsuperscript{42} (assumed to be aligned with the applied field \( H \)), and \( d \) is the film thickness.

For the \( r \)th PSSW mode, the wave vector \( q_n \) is quantized according to

\[
q_n = \frac{n \pi}{d},
\]

where \( n \) is an integer, and the observed frequency can be approximated as

\[
\omega_{\text{PSSW}} = \gamma \mu_0 \sqrt{\left( H + \frac{2A}{\mu_0 M_s} q_n^2 \right) \left( H + M_s + \frac{2A}{\mu_0 M_s} q_n^2 \right)},
\]

where the exchange constant \( A \) is introduced. These PSSW modes are thus rather insensitive to the angle of incidence.\textsuperscript{43}

Figure 6(b) shows the BLS spectra measured at various angles of incidence. The peaks near 26 GHz clearly shift position upon varying the angle of incidence, marking them as the DE modes. Conversely, the other observed peaks are assigned to the PSSW modes.

Using a model by Hillebrands\textsuperscript{44} based on a continuum-type magneto-static theory, the exchange constant \( A \), the related spin-wave stiffness \( D=[2g_g \mu_B]/[AM_s] \) (\( \mu_B \) being the Bohr magneton),\textsuperscript{25} the saturation magnetization, and the Landé \( g \)-factor can be determined from the experimentally observed spin-wave frequencies. Figure 7 shows the calculated dispersions for \( A=22.5 \pm 1.0 \, \text{pJ/m} \) (\( D = 413 \pm 15 \, \text{meV \AA}^2 \)), \( \mu_0 M_s=1.58 \pm 0.05 \, \text{T (6.43 \, \mu_B/\text{f.u.}} \), and \( g=2.0 \pm 0.1 \). As can be seen, these dispersions show a very good agreement with the observed spin-wave frequencies.

The exchange constant \( A \) we determine (22.5 pJ/m) is approximately twice as large as the values reported for Co\(_2\)MnGe(110) films grown on Al\(_2\)O\(_3\) by Belmeguenai \textit{et al.}\textsuperscript{27,28} In those studies, \( A \) was found to vary between 9.8 and 13.8 pJ/m for film thickness ranging from 30 to 83 nm. A discrepancy of this order is not surprising, as the exchange constant was shown to be dependent on the ordering\textsuperscript{19} and the number of valence electrons (i.e., composition).\textsuperscript{45} These two physical attributes are likely to be different between the samples studied by Belmeguenai and co-workers and the one studied here.

The effect of point defects on the half-metallicity and saturation magnetization have been studied by means of \textit{ab initio} calculations for Co\(_{x+y}\)Mn\(_{1-x}\)Ge (Refs. 46–49) and Co\(_{x+y}\)Mn\(_{1-x}\)Ge\(_{1+y}\) (Ref. 50) compounds. While Co\(_{2}\)Mn antisites, where a Mn site is replaced by a Co atom, destroy half-metallicity,\textsuperscript{46–48} this important feature is preserved for Mn\(_{2}\)Co antisites.\textsuperscript{47–49}

Slater–Pauling behavior relates the magnetic moment \( m \) per formula unit (or strictly speaking, per unit formula for a
nonstoichiometric material, including vacancies) to the number of valence electrons $N_v$ in the formula unit by the simple relationship:

$$m = N_v - 24.$$

This behavior is expected for half-metallic ferromagnets, and predicted to remain valid for $\text{Co}_2\text{Mn}_{1+x}\text{Ge}_{1-x}$ for $-0.2 < x < 0.2$. It is worth mentioning that this feature is also maintained for closely related Mn-rich $\text{Co}_2\text{MnSi}$ compounds. We will now discuss a possible formula unit composition of the $\text{Co}_2\text{Mn}_{0.77}\text{Ge}_{0.42}$ films based on this extended Slater–Pauling behavior as an approximation for the smaller than the $\text{Mn}$ are likely to be induced for Mn- and Ge-deficient $\text{Co}_2\text{MnSi}$ antisites are assumed and the Ge sites are approximately half-occupied, this tentative formula unit composition provides a Slater–Pauling behavior. Furthermore, while the theoretical prediction that a vacancy at a Si site is also maintained for closely related Mn-rich $\text{Co}_2\text{MnSi}$. The perturbation leading to the observed complex uniaxial anisotropy of the magnetization reversal processes (see Fig. 2) apparently has little influence on the electronic structure of the material, in particular, with respect to the anisotropy of the spin-orbit coupling (see Fig. 5 and related discussion).

**IV. CONCLUSION**

We have examined an epitaxial, nonstoichiometric Mn- and Ge-deficient $\text{Co}_2\text{Mn}_{0.77}\text{Ge}_{0.42}$ thin films by means of MOKE magnetometry and BLS spectroscopy. We find the sample exhibits a unidirectional anisotropy. A modest quadratic MOKE with an amplitude of 0.4 mdeg is observed. Using BLS spectroscopy, we find the exchange constant $A = 22.5$ J/m (exchange stiffness $D = 413$ meV Å$^2$), which is twice higher than previously reported for $\text{Co}_2\text{MnGe}(110)$ films. Even though the composition is highly off-stoichiometric, the magnetic moment appears to be approximated by Slater–Pauling behavior. Furthermore, while the magnetization reversal shows a clear uniaxial symmetry, the magneto-optical tensor describing this material is not significantly perturbed with respect to the ideal cubic symmetry.

**ACKNOWLEDGMENTS**

We thank Dr. J. Lösch (IFOS, TU Kaiserslautern) for x-ray measurements. We also thank Associate Professor T. Uemura (Hokkaido University) for helpful discussions. This project was financially supported by the DFG Research Unit 559, “New Materials with High Spin Polarization” (Germany). S.T. gratefully acknowledges the Alexander von Humboldt foundation for a postdoctoral research fellowship. This work was partly supported by a Grant-in-Aid for Scientific Research (A) (Grant No. 20246054), a Grant-in-Aid for Scientific Research on Priority Area “Creation and control of spin current” (Grant No. 19048001), and a Grant-in-aid for Hokkaido Innovation through Nanotechnology Support (HINTS) from the MEXT (Japan).