Letter to the Editor

Composition dependence of magnetic anisotropy and quadratic magnetooptical effect in epitaxial films of the Heusler alloy Co$_2$MnGe

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ABSTRACT

Magnetic anisotropy and magnetooptic Kerr effect for epitaxial films of Co$_2$Mn$_x$Ge$_{1-x-y}$ grown on Ge (111) substrates have been studied systematically in the compositional vicinity of the Heusler alloy Co$_2$MnGe. A large quadratic magnetooptic Kerr effect has been observed within a narrow region of composition centered around the Co to Mn atomic ratio of 2. The effect has been used to probe and quantify the magnetic anisotropy of the system, which is shown to have a strong sixfold in-plane component accompanied by a weak uniaxial component at room temperature. These properties are shown to depend sensitively on atomic ratio between Co and Mn, indicating the presence of an intrinsic composition-driven phenomenon.

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Heusler alloys belong to an interesting class of ternary compounds that includes many predicted half-metals [1,2]. Their spin-dependent states and magnetism are expected to depend sensitively on stoichiometry, epitaxial constraints, and the presence of various structural and chemical disorders [3], but very little is known about these dependencies, owing primarily to the complexity associated with a ternary system and thus the difficulties for studying these. For example, dependence of magnetic anisotropy on stoichiometry is an important and yet relatively unexplored property, critical for understanding and controlling the magnetism in this important class of materials. Recent advances in combinatorial molecular-beam epitaxy (MBE) have made it possible to map the composition of an entire ternary system onto a single substrate and to explore the material system systematically [4]. In this paper, we report a systematic study of magnetic anisotropy and magnetooptical properties of a ternary system Co$_{x}$Mn$_{y}$Ge$_{1-x-y}$ in the compositional vicinity of the Heusler alloy Co$_2$MnGe, a predicted half-metal with a high Curie temperature [2].

The ternary combinatorial epitaxial film was grown by MBE techniques on a Ge (111) substrate at 250 °C and at a deposition rate of 0.1 Å/s to a nominal thickness of ~630 Å. A multilayer method was employed via sequential deposition of submonolayer “wedges” of the 3 elements [5]. The alloy film was subsequently annealed at 450 °C. The growth and annealing conditions were optimized for the best structural quality of the Heusler composition [6]. The ternary composition was examined and quantified using an array of complementary techniques, including X-ray fluorescence spectroscopy [7], secondary ion mass-spectrometry, electron energy dispersive X-ray spectroscopy, and Rutherford backscattering spectroscopy. In Fig. 1(a), a schematic diagram of the combinatorial sample is shown, indicating the ternary region (the triangle) and the location of Co$_2$MnGe. Structural investigations using regular and anomalous X-ray diffraction techniques [7,8] indicate that epitaxial films within this region of composition exhibit a high degree of structural and chemical ordering. The magnetooptical Kerr effect (MOKE) measurements were performed at room temperature in the longitudinal geometry using a diode laser (wavelength of 664.3 nm). A standard lock-in technique with a photoelastic modulator was used for simultaneous detection of both Kerr rotation and ellipticity. The laser spot on the sample was focused to ~100 μm in diameter, which corresponds to ~1 at% in the composition space. The sample was mounted on a precision translation and rotation stage, and it was scanned with respect to the laser spot in order to probe the composition dependence. MOKE hysteresis loops were measured at each position with the magnetic field directed along more than 10 in-plane directions, including all of the in-plane $\langle 110 \rangle$ and $\langle 112 \rangle$ directions. Scribed crosshairs with line width of ~10 μm on the sample were used to position the laser spot on the sample, resulting in a positional reproducibility of better than 20 μm (~0.2 at% in composition) between different measurements.

MOKE hysteresis loops in the vicinity of Co$_2$MnGe exhibit a pronounced asymmetry with respect to the origin, as shown in Fig. 1(b) and (c). For example, as the Ge concentration increases through the Heusler stoichiometry, a characteristic “spike” emerges in the hysteresis loop near the positive coercive field (Fig. 1(c)); at a fixed Ge concentration, as the atomic ratio between Co and Mn passes through the value of 2, a reversal of the
The consequence is that both magnetization projections parallel and perpendicular to the field, $M$ and $J$, respectively, are detected by MOKE. For instance, a sudden jump in magnetization direction can produce a "spike" in the hysteresis loop, where a high degree of structural and chemical ordering has been observed [8]. The QMOKE values reported here are comparable to previously reported values for Heusler alloys of Co$_2$FeSi [11] and PtMnSb [12]. Since large QMOKE values enhance the sensitivity for detecting and quantifying magnetization directions (Eq. (1)), the effect has been used to analyze the magnetic anisotropy of the system, as discussed below.

The measured MOKE hysteresis loops as a function of field directions have been simulated using Eq. (1). The magnetization directions, on the other hand, have been calculated using a single domain Stoner–Wohlfarth model [13,14] by minimizing the energy density associated with the in-plane magnetic anisotropy, $E_{\text{IPMA}}$. For the (111) symmetry, the energy density is given by

$$E_{\text{IPMA}} = K_u(28 - \cos \theta)/108 + K_6 \sin^2(\theta - \varphi) - M_s H.$$  

Here, the sixfold magnetic anisotropy with the corresponding constant $K_u$ (the first term) arises from crystal symmetry in thin-film geometry, with $\theta$ being the angle between the direction of magnetization and one of the sixfold easy axes (for $K_u > 0$). A uniaxial correction ($K_6$) is added (the second term) with $\varphi$ the angle between the uniaxial easy axis and one of the sixfold easy axes. The last term represents the Zeeman energy.

The model simulations, as described above using $K_6/M_s$, $K_u/M_s$, $Q_0/L$, and $Q_1/L$ as the adjustable parameters, have yielded good qualitative agreements with the measured hysteresis loops, as shown in Fig. 3 for a typical set with $K_6/M_s = (1200 \pm 100)$Oe and $K_u/M_s = (20 \pm 3)$Oe. The sixfold easy axes are determined to be along the in-plane $\langle 110 \rangle$ directions consistent with the crystal symmetry. A weak uniaxial magnetic anisotropy (UMA) is often needed in order to produce the distinct features in the hysteresis loops, and near the Heusler stoichiometry, the easy axis for UMA is near [011]. We note that the observed magneto-optical parameters at a single composition depend on the geometries of the measurement. Specifically, the parameters $Q_0$ and $Q_1$ exhibit a monotonic increase with decreasing incident angle, while the counterpart $L$ exhibits a corresponding decrease [9,10,15]. In addition, a second-order magnetization-induced...
Quantitative values of magnetic anisotropy as a function of composition dependence, particularly the symmetry and sensitivity on the Co to Mn atomic ratio, strongly support the presence of an intrinsic composition driven phenomenon. Such a phenomenon is not previously known for any Heusler alloys, and it appears to be responsible for the observed ordering around Co/Mn ratio of 2 and for the observed strong correlation and interplay between structure, magnetism, and magnetooptical effects. Since both magnetic anisotropy and magnetooptical effects, in particular QMOKE, are the result of corresponding spin-orbit interactions, the electronic structure of the system and perhaps chemical ordering within the lattice may also be driven by the same composition dependent phenomenon. These findings should provide the necessary impetus for future work in this material system in order to elucidate the origin of the composition driven effect and to control magnetism and spin-dependent states. From a broader perspective, while properties of complex alloys are generally expected to depend on composition, investigations into how the dependence takes place should still take precedence.

While there are various extrinsic sources and interactions that affect the nature and strength of magnetic anisotropy and magnetooptical effect for any given sample, the observed composition dependence, particularly the symmetry and sensitivity on the Co to Mn atomic ratio, strongly support the presence of an intrinsic composition driven phenomenon. Such a phenomenon is not previously known for any Heusler alloys, and it appears to be responsible for the observed ordering around Co/Mn ratio of 2 and for the observed strong correlation and interplay between structure, magnetism, and magnetooptical effects. Since both magnetic anisotropy and magnetooptical effects, in particular QMOKE, are the result of corresponding spin-orbit interactions, the electronic structure of the system and perhaps chemical ordering within the lattice may also be driven by the same composition dependent phenomenon. These findings should provide the necessary impetus for future work in this material system in order to elucidate the origin of the composition driven effect and to control magnetism and spin-dependent states. From a broader perspective, while properties of complex alloys are generally expected to depend on composition, investigations into how the dependence takes place should still take precedence.

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References