Development of Co-Mn-Al thin films with giant anomalous Hall effect towards read head applications

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Recent advancements in magnetic recording densities have led to a growing demand for a new read head technologies achieving sufficient SNR in a small sensor size. Anomalous Hall effect (AHE) sensors can be a suitable candidate for the next generation read heads. In this work we explore the composition dependence of AHE in Co-Mn-Al (CMA) Heusler alloys for a wide range of compositions within a small deposition area. For this, we employed a high-throughput technique based on composition gradient films fabricated by combinatorial sputtering. The AHE results from this two-dimensional (2D) composition-spread film provided a critical range of Co, Mn and Al compositions within which one can yield the best AHE properties. This experiment demonstrated that Al-rich off-stoichiometric compositions provided higher anomalous Hall resistivity (ρ_{yx}) than the stoichiometric one. In order to have a detailed material investigation and to study the post-annealing temperature (T_A) dependence, uniform films with composition based on the above results were prepared. The highest ever reported value of ρ_{yx} (~ 30 $\mu\Omega$ ·cm) for films post-annealed at 500 °C has been achieved on a Si/SiO_x substrate. The results show that CMA Heusler alloys are potential candidates for application in AHE based reader devices.

Index Terms—Anomalous Hall effect, magnetic read-head, Heusler alloys, combinatorial sputtering, composition-spread films.

I. INTRODUCTION

THE ever-increasing demand for higher areal density in hard I disk drives (HDDs) necessitates the development of advanced magnetic sensor materials for read head devices with improved reading resolutions and signal-to-noise ratio [1]. AHE-based reader devises have been identified as promising candidates for the next-generation read head technologies due to their high linearity, larger dynamic range, high output voltage, and predicted low noise level [2]. A schematic diagram showing an AHE-reader for reading the magnetization information from magnetic bits is shown in Fig. 1a. The schematic of currently used TMR reader and a proposed structure for AHE reader are shown in Fig. 1c and 1d. The AHE reader has a simpler structure with a single sensing layer. Thicker sensing layers can be used within the allowable gap space, unlike the free layers in TMR readers. This increased thickness helps suppress magnetization noise caused by thermal fluctuations, which is a major issue in miniaturized readers. In [2] the output voltages for AHE-based sensing layer (SL) and a TMR-free layer (FL) in response to an external field (analogous to a media field from magnetic bits) is estimated based on micromagnetic simulations (see Fig. 1b). The output voltage in the case of TMR is clearly assymetric in comparison to AHE. Higher linearity and improved dynamic range is also attained for AHE based SL. The requirement of materials with large AHE, from an application viewpoint to overcome the issues such as bias current shunting and output voltage leakage is also pointed out in this work. A giant roomtemperature AHE with ρ_{yx} of 36.9 $\mu\Omega$ cm has been reported in bulk single crystal Co₂MnAl [3]. For epitaxial Co₂MnAl thin films deposited by combinatorial sputtering, ρ_{yx} up to ~22 $\mu\Omega$.cm has been reported [4].

It is known that one of the dominant mechanisms for large AHE arises from the electronic band structure of the material, particularly, the presence of large Berry curvature near the Fermi level [5]. As the position of the Fermi level is sensitive to changes in stoichiometry, composition variation is expected

to have a significant effect on the AHE properties. However, a comprehensive study on the effect of composition on ρ_{yx} for polycrystalline CMA films is lacking. Therefore, in this work, we studied the composition dependence of AHE in CMA by



Fig. 1. (a) Schematic of an AHE reader. (b) comparison of output voltage for AHE and TMR readers. (c) Schematic of the existing TMR reader structure (d) Proposed AHE reader structure. W is reader width, and G is read gap. The white arrows in the shields and free (sensing) layer indicate the magnetization directions of the layers. (Data from [2])

using a high-throughput technique based on combinatorial sputtering.

II. EXPERIMENTAL METHODOLOGY

A. Composition-spread films

Two-dimensional (2D) composition-spread thin films of CMA were fabricated on $10 \times 10 \text{ mm}^2$ thermally oxidized Si (Si/SiO_x) substrates using a combinatorial sputtering system (CMS-A6250X2, Comet, Inc.). The deposition was performed at room temperature and an Argon gas pressure of 0.6 Pa. As shown in Fig. 1a, composition-spread region forms a triangular area with the three vertices designed to have compositions of Co₆₂Mn₁₆Al₂₂, Co₄₂Mn₁₆Al₄₂ and Co₄₂Mn₃₆Al₂₂. The deposition was carried out by co-sputtering from Co, Mn and Al targets. Initially, uniform layers of Co, Mn and Al of thickness 0.17 nm,

0.074 nm and 0.142 nm, respectively, are deposited, and subsequently, wedge-shaped layers with thickness varying from 0 to 0.114 nm are deposited sequentially for each of the elements. After depositing a wedge for each element, the substrate was rotated by 120° before the wedge deposition of the subsequent layer. The deposition cycle of uniform and wedge layers is repeated for 60 times to form a total thickness of ~25 nm in the triangle region. The deposited films are then in-situ post-annealed at 500 °C for 30 minutes. A Ta capping of 3 nm was deposited after the film is cooled down to room temperature.

Hall-bar devices of length and width 80 and 20 μ m, respectively, were fabricated on the 2D composition-spread film for AHE measurements. Because of the small dimensions, the composition gradient within the individual Hall-bar devices is negligibly small. AHE and longitudinal resistivity (ρ_{xx}) measurements are conducted using a Physical Property Measurement System (VersaLab, Quantum Design).

The compositions of individual Hall-bar devices were measured for selected devices using electron probe microanalysis (EPMA). The composition for the rest of the devices were estimated by interpolating from the measured composition values. A slight deviation from the designed composition values was observed, and EPMA-based values are used in the following part.

B. Uniform films

Uniform films of CMA with a thickness of 30 nm were fabricated by co-sputtering from $Co_{66}Mn_{34}$ and Al targets. The films are capped with a 3 nm SiO₂ layer. Ex-situ post-annealing of the deposited films was performed at 500 °C for 1 h. The composition measurement was conducted by X-ray fluorescence spectroscopy (XRF) using a quantitative analysis method, which was calibrated using standard Co-Mn-Al films whose compositions were premeasured by inductively coupled plasma mass spectrometry.



Fig. 2. Ternary contour plot showing the variation of ρ_{yx} with Co-Mn-Al composition.

III. RESULTS

Figure 2 shows the AHE results for the 2D compositionspread film. The ternary contour plot depicts the variation of ρ_{yx} with CMA composition, based on the data obtained from 35 Hall-bar devices (black dots). A clear compositional dependence of ρ_{yx} can be seen. For a composition close to the stoichiometric one (Co₅₀Mn_{24.5}Al_{25.5}), ρ_{yx} was ~17 $\mu\Omega$ ·cm, which is consistent with the previously reported value [6].

The ρ_{yx} values over 26 $\mu\Omega$ ·cm, represented by the red region in Fig. 2, were obtained for off-stoichiometric Al-rich compositions with the highest value of ~27.7 $\mu\Omega$ ·cm achieved at Co_{45.1}Mn_{20.4}Al_{34.5}. Scanning transmission electron microscopy (STEM) observation of a device with composition Co_{46.3}Mn_{20.7}Al₃₃ and a high ρ_{yx} (26.9 $\mu\Omega$ ·cm) showed that the film had a polycrystalline structure with *B*2 atomic ordering.



Fig. 3. (a) AHE behavior for uniform-CMA films with varying compositions. (b) ρ_{yx} dependence on post-annealing temperature for $Co_{47.9}Mn_{21.2}Al_{30.9}$.

AHE results for uniform CMA films annealed at $T_A = 500$ °C with varying Al concentrations from 28.3 to 34.3 at.% is shown in Fig. 3a. The saturation field, which represents the saturation magnetization due to demagnetization field, decreases with increasing Al. The highest ρ_{yx} of 30.4 $\mu\Omega$ ·cm is obtained for Co_{47.9}Mn_{21.2}Al_{30.9}. The annealing temperature dependence of this film exhibits a monotonic increase in ρ_{yx} up to $T_A = 500$ °C and showed no improvement in ρ_{yx} with increasing T_A .

IV. CONCLUSION

In conclusion, we have employed a high-throughput combinatorial sputtering technique to explore the composition dependence of the anomalous Hall effect (AHE) in CMA. Based on this result, uniform films with the highest ever reported value of ρ_{yx} of 30.4 $\mu\Omega$ ·cm were achieved for Co_{47.9}Mn_{21.2}Al_{30.9} on polycrystalline substrates. Such high values, along with linear response and large dynamic range, are advantageous for applications in magnetic reader devices. The temperature dependence of these films was also studied.

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