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## Oblique angle deposition-induced anisotropy in Co<sub>2</sub>FeAl films

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#### ABSTRACT

A series of  $Co_2$ FeAl Heusler alloy films, fabricated on  $Si/SiO_2$  substrates by magnetron sputtering-oblique angle deposition technique, have been investigated by magnetization and transport measurements. The morphology and magnetic anisotropy of the films strongly depended on the deposition angle. While the film deposited at zero degree (i.e. normal incidence) did not show any anisotropy, the films deposited at higher angles showed unusually strong in-plane anisotropy that increased with deposition angle. The enhanced anisotropy was well-reflected in the direction-dependent magnetization and the coercivity of the films that increased dramatically from 30 Oe to 490 Oe. In a similar vein, the electrical resistivity of the films also increased drastically, especially for deposition angles larger than  $60^\circ$ . These anisotropic effects and their relation to the morphology of the films are discussed.

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#### 1. Introduction

From both fundamental and applied points of view, the Heusler class of intermetallic alloys have been the subject of intense research interest, since their high spin polarization makes them candidate materials for spintronic devices. [1,2]  $Co_2FeAl$  (CFA) is one of the most attractive Heusler alloys due to its high Curie temperature (Tc = 1000 K) [3], relatively low magnetic damping parameter (around 0.001) [4], and high spin polarization. Because of these traits, CFA has been identified for potential use as an alternative magnetic electrode material in data storage architectures such as magnetic tunnel junctions and current-perpendicular-to-plane (CPP) spin valves. CFA/MgO/CoFe magnetic tunnel junctions have been shown to possess an immense tunneling magnetoresistance (up to 360%) at room temperature [5], further fueling research interest in CFA.

CFA films (thickness ranging from 10 nm to 100 nm) grown under standard conditions on Si and MgO substrates typically exhibit small in-plane coercivity ( $H_c$  < 50 Oe). [6,7] Recently, Niesen et al. showed that TiN buffered ultrathin ( $\sim$ 1 nm) CFA/MgO bilayer films exhibit  $H_c$  of as large as  $\sim$  320 Oe. [8] Li et al. showed that CFA films fabricated by cosputtering with Tb exhibit significantly large  $H_c$  of 800 Oe. [9] The large  $H_c$  of these ultrathin CFA films are generally associated with their perpendicular-to-plane magnetic anisotropy. These experimental results (along with many others, reported in literature) suggest that the magnetic anisotropy of

A fabrication technique by which uniaxial magnetic anisotropy can be introduced in thin films is oblique angle deposition (OAD) [13–16]. The technique is also known as glancing angle deposition (GLAD). [17] In this technique the deposition flux approaches a stationary substrate at an angle  $\alpha$  relative to the substrate normal, referred to as the incidence angle. Due to atomic shadowing effects [18], tilted nano-columnar structures are formed which exhibit properties in severe contrast to those of thin films prepared with the deposition normal to the substrate. Besides magnetic anisotropy, these nano-columnar structures possess a higher surfacearea-to-volume ratio, which can have a dramatic impact on the electronic transport [19–21] and optical properties [21,22]. The significant changes in physical properties introduced by OAD are relevant to a variety of data storage and sensing applications.

Up-to-date, no report has been made of CFA films deposited by OAD technique. Considering the fact that the magnetic anisotropy of these films can be controlled dramatically by fabrication conditions, it is interesting to study OAD thin films of CFA. Therefore, in this article we report on the magnetic and electrical resistivity properties of CFA thin films prepared by magnetron-sputtering OAD (MS-OAD). A significant enhancement of uniaxial anisotropy has been observed in the films due to OAD.

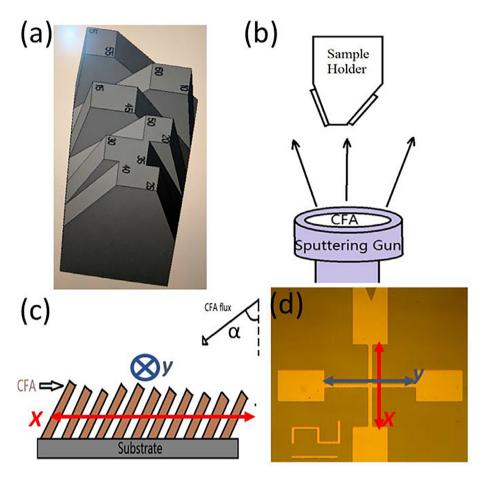
#### 2. Experiment

To allow for the simultaneous growth of several CFA films at different incidence angles, a plastic block (Fig. 1a) consisting of chiseled surfaces with tilt angles ranging from  $30^{\circ}$  to  $85^{\circ}$  at  $5^{\circ}$ 

CFA films can be controlled by manipulating their thickness and fabrication conditions [10-12].

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**Fig. 1.** (a) Diagram of the sample holder design showing various tilted surfaces. Six planes with distinct tilt angles were fabricated on each of the two long sides of the holder. The oblique incidence angle  $\alpha$  of the left side surfaces, from top to bottom, was 85°, 30°, 75°, 40°, 60°, and 50°. Similarly, the incidence angle  $\alpha$  of the right side surfaces was 35°, 80°, 45°, 70°, 55°, and 65°. (b) Schematic view of MS-OAD (vertical distance not to scale). (c) Cartoon showing the nano-pillar mode of growth in OAD. The coordinate convention employed in the report is also indicated. (d) Image of a microstructure used for resistivity measurement. The length and the width of the thin channels are 400 μm and 27 μm respectively. The small wedge shape on the top rectangular extension pad marks the direction of the in-plane projection of the CFA flux.

intervals was fabricated using a 3-D printer. Fig. 1(b) is a schematic diagram of the MS-OAD setup employed, consisting of a 1 in. sputtering gun with the sample holder 20 cm above. Clean SiO<sub>2</sub>-coated Si substrates (1000 nm SiO<sub>2</sub>) with approximate dimensions of  $0.5 \text{ cm} \times 1 \text{ cm}$  were mounted on the angled surfaces depicted in Fig. 1(a). The base pressure of the deposition chamber was  ${\sim}2{\times}~10^{-7}$  mbar and the CFA films were sputtered at room temperature under a  $3 \times 10^{-3}$  mbar partial Ar pressure. CFA was deposited on the substrates at a rate of about 0.17 Å/s, as determined by a quartz crystal monitor. Two in-plane directions were chosen for anisotropy measurements: One direction parallel to the projection of the CFA flux on to the plane of the substrates (henceforth referred to as the parallel direction or x-axis), and the other perpendicular to the projection (henceforth referred to as the y-axis), as shown in Fig. 1(c). Non-patterned thin films were used for magnetization measurements, while photolithographic techniques were utilized to create cross-shaped micro-channels for one batch of substrates to be used in anisotropic measurements of the electrical resistivity, as depicted in Fig. 1(d). These micro-channels were mounted in the holder such that one set of channels was along the previouslydefined x-axis and the other was along the y-axis (i.e. the channel sets were perpendicular to each other). The morphological characteristics of the films were assessed using a Zeiss Supra 35 VP FEG SEM, using the in-lens secondary electron detector. Elemental quantification was provided using a Bruker Quantax energy-dispersive X-ray spectroscopy (EDS) unit housed within the SEM. The static magnetic properties were determined using the vibrating sample magnetometer (VSM) option of a Quantum Design Physical Property Measurement System (PPMS), in applied fields up to 1 kOe in strength. The room temperature electronic transport properties were probed using the four-probe resistivity module of the PPMS. The thickness of the micro-channels was measured using a Nanosurf easyScan 2 atomic force microscope (AFM).

#### 3. Results and discussions

SEM images of films deposited with  $\alpha$  = 0° and 85° are shown in Fig. 2(a) and (b), respectively. From these images, it is clear that while the grains form a tight pattern on the normally-deposited ( $\alpha$  = 0°) film, the grains on the surface of the OAD-prepared films are more discrete. In the sample fabricated with  $\alpha$  = 85°, there exist gap intervals between grains, evidenced by the darker areas in Fig. 2b. These intervals extend in the *y*-direction, indicating that columns have a greater chance of being in contact with other columns along the *y*-direction than being in contact with columns along the *x*-direction. Furthermore, the surface of the normally-deposited film appears smoother than the film deposited with  $\alpha$  = 85°. EDS analysis of the CFA layer rendered elemental quantifications matching the target stoichiometry.

Fig. 3a–c show the magnetization as function of applied magnetic field [M(H)] curves, collected under the zero-field cooling protocol (ZFC) at 10 K, for the samples deposited at  $\alpha = 0^{\circ}$ , 40°, and 70°, respectively. For each sample, the magnetization measured at each temperature M has been normalized to the saturation magnetization  $M_S$ . For the normally-deposited sample ( $\alpha = 0^{\circ}$ ), M

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