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Magnetic and structural properties of Co₂FeAl thin films grown on Si substrate

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The correlation between magnetic and structural properties of Co₂FeAl (CFA) thin films of different thickness (10 nm < d < 100 nm) grown at room temperature on MgO-buffered Si/SiO₂ substrates and annealed at 600°C has been studied. XRD measurements revealed an (011) out-of-plane texture growth of the films. The deduced lattice parameter increases with the film thickness. Moreover, pole figures showed no in-plane preferential growth orientation. The magneto-optical Kerr effect hysteresis loops showed the presence of a weak in-plane uniaxial anisotropy with a random easy axis direction. The coercive field measured with an applied field along the easy axis direction and the uniaxial anisotropy field increase linearly with the inverse of the CFA thickness. The microstrip line ferromagnetic resonance measurements for in-plane and perpendicular applied magnetic fields revealed that the effective magnetization and the uniaxial in-plane anisotropy field follow a linear variation versus the inverse CFA thickness. This allows deriving a perpendicular surface anisotropy coefficient of -1.86 erg/cm².

Keywords: Heusler alloys ; Magnetic anisotropy; Ferromagnetic resonance; Magnetization dynamics; Heusler alloys

I. INTRODUCTION

Heuslers X_2YZ (X being a transition metal element, Y being another transition metal element and Z being a group *III*, *IV*, or *V* element) are interesting group of materials due to their potential use as one magnetic electrode in giant and tunnel magnetoresistance devices used in magnetic memories (MRAM) [3], in low field magnetic sensors [4] and in microwave components for spintronic applications [5]. The most prominent representatives of the this kind of spintronic materials are Co-based full Heusler alloys due to their predicted half metallic character (magnetic materials that exhibit a 100% spin polarization at the Fermi level) even at room temperature and due to their high Curie temperature [6]. These make them ideal sources of high spin polarized currents to realize high giant magnetoresistance values. Co₂FeAl (CFA) is one of the Co-based Heusler alloys having a very high Curie temperature (1000 K) and, theoretically predicted, a half-metallic character of their spin-split band structure. It can provide giant tunnelling magnetoresistance (360% at room temperature) [7] when used as an electrode in magnetic tunnel junctions, which makes CFA promising for practical applications. However, the crystalline structure and the chemical order of such materials strongly influence their magnetic and structural properties. Moreover, the substrate material as well as the orientation of substrate and the film thickness have

an impact on the magnetic anisotropy of magnetic thin films such as CFA because of the band hybridization and the spin-orbit interaction at the interface. Therefore, the correlation between their magnetic and structural properties and its dependence on film thicknesses, for precise control of the magnetic properties required by the integration of CFA as a magnetic electrode in spintronic devices, should be investigated. For this purpose, microstrip ferromagnetic resonance (MS-FMR), magneto-optical Kerr effect (MOKE) magnetometry and X-ray diffraction (XRD) techniques are used.

II. SAMPLE PREPARATION

CFA films thin films of different thickness (10 nm < d < 100 nm) were grown on Si(001)/SiO₂ substrates using a magnetron sputtering system with a base pressure lower than 3×10^{-9} Torr. Prior to the deposition of the CFA films, a 4 nm thick MgO buffer layer was grown at room temperature (RT) by rf sputtering from a MgO polycrystalline target under an Argon pressure of 15 mTorr. Next, CFA thin films of different thickness, were deposited at room temperature by dc sputtering under an Argon pressure of 1 mTorr, at a rate of 0.1 nm/s. Finally, the CFA films were capped with a MgO(4 nm)/Ta(4 nm) bilayer. After the growth of the stack, the structures were ex-situ annealed at 600°C during 15 minutes in vacuum. The structural properties of the samples have been characterized by XRD using a four-circle diffractometer. Their magnetic static and dynamic properties have been studied by magneto-optical Kerr effect magnetometer (MOKE) and microstrip ferro-

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magnetic resonance (MS-FMR) [8], respectively.

III. STRUCTURAL PROPERTIES

The X-rays $\theta - 2\theta$ diffraction patterns for CFA thin films of different thicknesses revealed one peak which is attributed to the (022) diffraction line of CFA (Fig. 1a). The lattice parameter (a), shown on Figure 1b, increases with the increasing CFA thickness, similarly to samples grown on MgO substrates where a direct correlation exists with the enhancement of the chemical order [9], however the lattice parameters remain smaller than the reported one in the bulk compound with the L2₁ structure

(0.574 nm). Due to the overlap of the potential (002) CFA peak with the substrate reflections it is difficult to accurately evaluate the degree of chemical order in our films. Pole figures around the (022) type CFA peaks (Fig. 1c) indicates that the CFA films show a strong (011) fiber- texture with no in-plane preferential growth direction.

IV. MAGNETIC PROPERTIES

To analyze the experimental results, the total magnetic energy given by equation (1) is considered.

$$E = -M_S H (\cos(\varphi_M - \varphi_H) \sin \theta_M \sin \theta_H + \cos \theta_M \cos \theta_H) - (2\pi M_S^2 - K_{\perp}) \sin^2 \theta_M - \frac{1}{2}(1 + \cos(2(\varphi_M - \varphi_u)))K_u \quad (1)$$

In the above expression φ_M (resp. φ_H) represents the in-plane (referring to the substrate edges) angle defining the direction of the magnetization M_S (resp. the external applied field H), while θ_M (resp. θ_H) is the out-of-plane angle between the magnetization (resp. the external field H) and the normal to the sample plane. φ_u defines the angles of an easy uniaxial planar axis with respect to this

substrate edge. K_u , and K_{\perp} are in-plane uniaxial and out-of-plane uniaxial anisotropy constants, respectively.

For an in-plane applied magnetic field, the studied model provides the following expression (2) for the frequencies of the experimentally observable magnetic modes:

$$F_n^2 = \left(\frac{\gamma}{2\pi}\right)^2 \left(H \cos(\varphi_M - \varphi_H) + H_u \cos(\varphi_M - \varphi_u) + \frac{2A_{ex}}{M_s} \left(\frac{n\pi}{d}\right)^2 \right) \left(H \cos(\varphi_M - \varphi_H) + \frac{H_u}{2} (1 + \cos(\varphi_M - \varphi_u)) + \frac{2A_{ex}}{M_s} \left(\frac{n\pi}{d}\right)^2 + 4\pi M_{eff} \right) \quad (2)$$

where $\frac{\gamma}{2\pi} = g \times 1.397 \times 10^6$ Hz/Oe is the gyromagnetic factor. We introduce the effective magnetization $M_{eff} = H_{eff}/4\pi$ obtained by:

$$4\pi M_{eff} = H_{eff} = 4\pi M_S - \frac{2K_{\perp}}{M_S} = 4\pi M_S - H_{\perp} \quad (3)$$

As experimentally observed, the effective perpendicular anisotropy term K_{\perp} (and, consequently, the effective perpendicular anisotropy field H_{\perp}) is thickness dependent: K_{\perp} describes an effective perpendicular anisotropy term which writes as:

$$K_{\perp} = K_{\perp}^V + \frac{2K_{\perp}^S}{d} \quad (4)$$

where K_{\perp}^S refers to the perpendicular anisotropy term of the interfacial energy density. Finally we define $H_u =$

$2K_u/M_S$ as the in-plane uniaxial anisotropy field. The uniform precession mode corresponds to $n = 0$. The other modes to be considered (perpendicular standing modes: PSSW) are connected to integer values of n : their frequencies depend upon the exchange stiffness constant A_{ex} and upon the film thickness d .

In the case of an out-of-plane perpendicular applied magnetic field, the resonance frequency is given by:

$$F_{\perp} = \left(\frac{\gamma}{2\pi}\right) \left(H - 4\pi M_{eff} + \frac{2A_{ex}}{M_s} \left(\frac{n\pi}{d}\right)^2 \right) \quad (5)$$

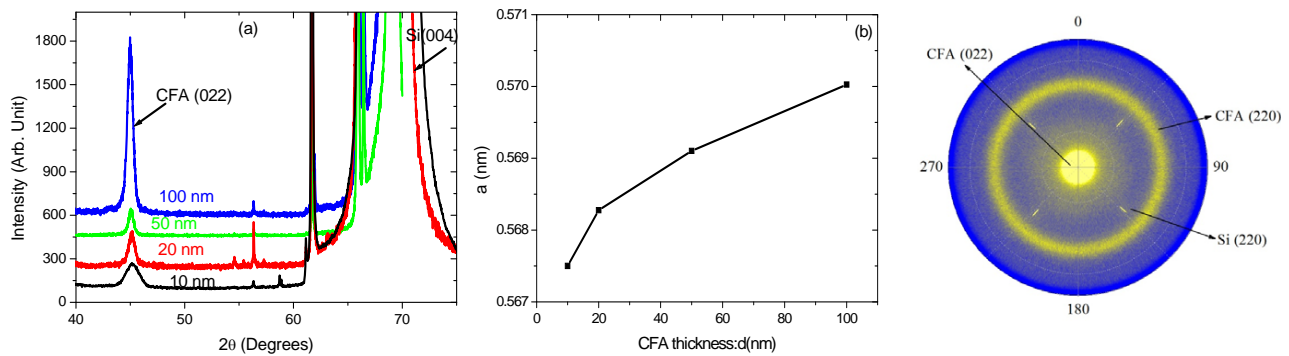


Figure 1: Thickness dependence of (a) $\theta - 2\theta$ patterns (b) lattice parameter; (c) pole figure around a (022) peak of 50 nm thick CFA film

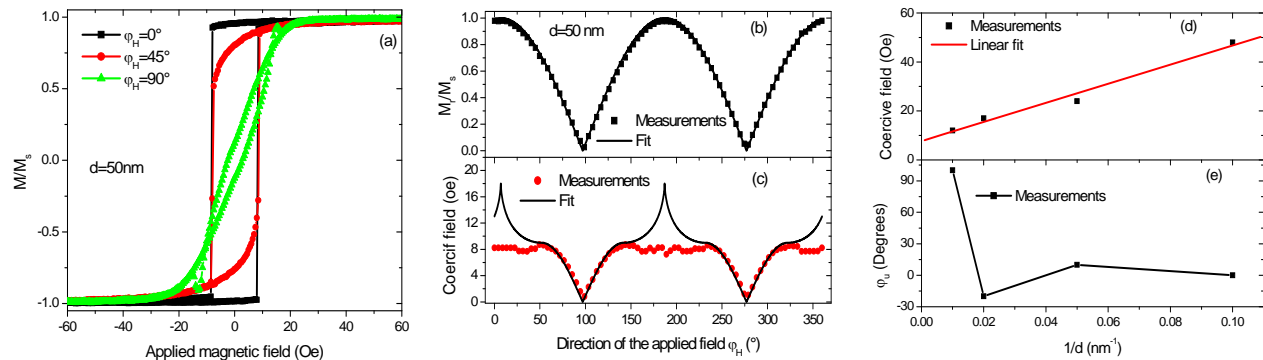


Figure 2: (a) MOKE hysteresis loops and the corresponding angular of the (b) normalized remanent magnetization (c) the coercive field of the 50 nm thick Co₂FeAl thin film. (d) and (e) thickness dependence of the easy coercive field and the magnetization easy axis respectively of the Co₂FeAl thin films.

1. Static properties

The overage magnetization at saturation measured by vibrating sample magnetometer at room temperature for all samples has been found to be $M_s = 1000 \pm 50$ emu/cm³. The typical MOKE hysteresis loops, measured versus external magnetic field orientations with respect to one of the substrate edges for all the samples, are represented on figure 2a for the 50 nm thick film. The shape of the magnetization reversal loops changes with the magnetic field orientation mainly due to the uniaxial magnetic anisotropy. The corresponding typical angular dependence of coercive fields (H_C) and normalized remanent magnetizations (M_r/M_s) are represented on figures 2b and 2c for the 50 nm sample. The samples have a similar angular dependence of H_C and M_r/M_s however, the H_C magnitude and the easy axes directions are different from each other (Fig. 2d and 2e). Apparently, the angular dependences agree with those of Stoner Wohlfarth (coherent rotation: CR) model as shown on figures 2b and 2c. Despite of the perfect agreement for M_r/M_s , a significant discrepancy for H_C is shown around the easy axis direction between the CR model and measurements. In fact, the coercive fields deduced from the magneti-

zation loops around the easy axis are smaller than the one obtained from CR model since that they are usually determined by domain nucleation and sample imperfection. The coercive field deduced from hysteresis loops obtained for a magnetic field applied along the direction of the easy axis, are shown in figure 2d as a function of the inverse film thickness. H_C increases linearly with the inverse of the film thickness due maybe to the enhancement chemical order or to the strain relaxation as the thickness increases.

2. Dynamic properties

The MS-FMR spectra measured for in-plane and perpendicular applied magnetic fields show the existence of a uniform mode for all samples and a first perpendicular standing spin wave mode only for the thickest films (100 and 50 nm thick films). Typical field dependencies of resonance frequencies of the uniform and the first PSSW mode are shown in figures 3a and 3b respectively for magnetic fields applied in-plane and perpendicular to the CFA film planes of various thicknesses. By fitting the data in figure 3 to the above presented model,

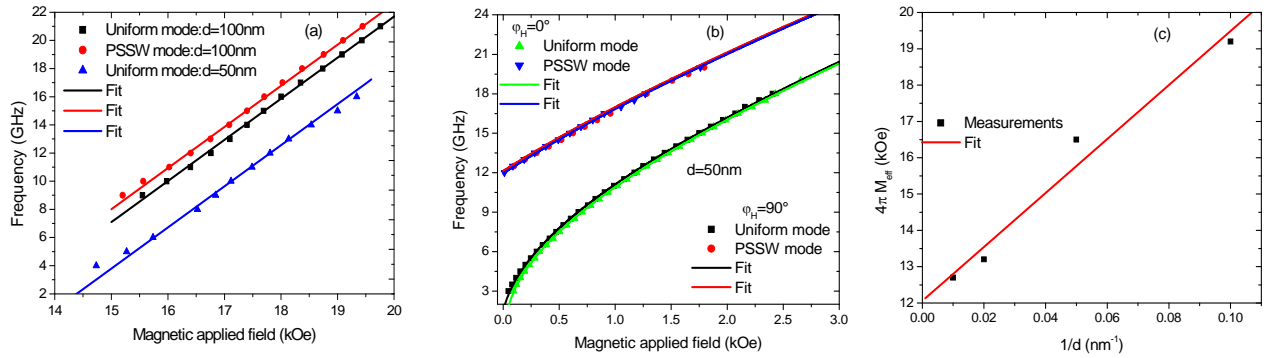


Figure 3: Uniform precession and perpendicular standing spin waves (PSSW) modes frequencies versus the (a) perpendicular and (b) in-plane applied magnetic fields for 100 and 50 nm thick Co₂FeAl thin films. The solid lines refer to the fit using the above mentioned model. (c) Thickness dependence of the effective magnetization, of Co₂FeAl thin films, deduced from the MS-FMR measurements

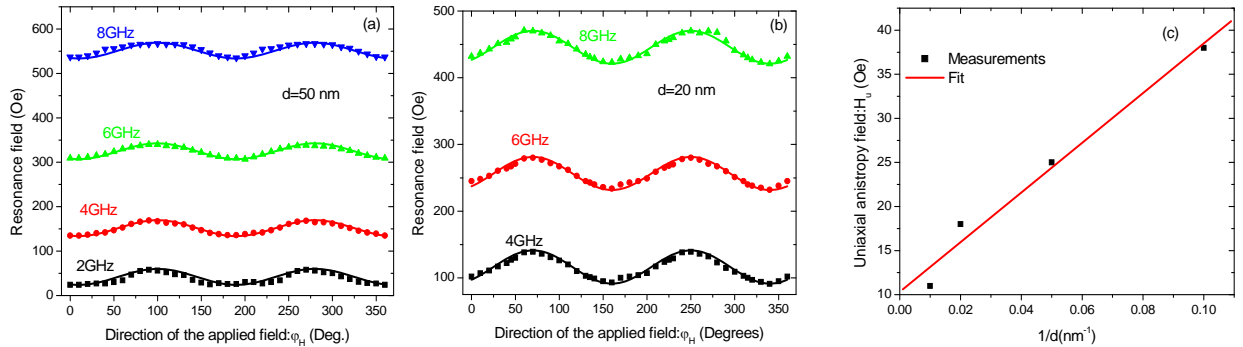


Figure 4: Angular dependence of the resonance field of (a) 50 nm and (b) 20 nm thick Co₂FeAl thin films. The solid lines refer to the fit using the above mentioned models. (c) Thickness dependence of the uniaxial (H_u) extracted from the fit of MS-FMR measurements. The solid lines are the linear fits.

the gyromagnetic factor (γ), exchange stiffness constant (A_{ex}) and the effective magnetization ($4\pi M_{eff}$) are extracted. The fitted $\gamma/2\pi$ and A_{ex} values are found to be a constant of 29.2 GHz/T and 1.5 μ erg/cm, respectively, across different samples. This exchange constant value is in good agreement with that indicated by Trudel et al. [6].

Interestingly, the extracted effective magnetization from the MS-FMR measurements, shown on figure 3c follows a linear variation versus the inverse CFA thickness. The linear fit of the measurements allows to determine the value of the perpendicular surface anisotropy constant: $K_{\perp}^S = -1.86$ erg/cm². The $4\pi M_{eff}$ value when d tends to infinity, equal to 12.05 kOe, is very close to the M_s mentioned above, taking into account experimental accuracy. This value of the surface perpendicular anisotropy constant is in the range (1.3 erg/cm² [10] and 1.8 erg/cm² [11]), but of an opposite sign with those reported in the MgO-based magnetic tunnel junctions having CoFeB electrodes or in Ta/CFA/MgO multilayers [12]. Ikeda et al. [10] attributed this perpendicular anisotropy in the Ta/CoFeB/MgO structure to

the contribution of the CoFeB/MgO interface. Wang et al. [13] have also argued that this anisotropy is due to the CoFeB/MgO interface based on the fact that it is only observed when the MgO thickness overpasses 1 nm. Therefore, we conclude that the uniaxial perpendicular anisotropy derives from surface energy term. Its negative value favors in-plane orientation of the magnetization. The origin of this anisotropy is most likely due to the CFA/MgO interface. This was confirmed (note shown here) by measuring the effective magnetization of a 10 nm CFA thick layer deposited on a SrTiO₃ substrate by using a 3 nm CoFe thick buffer layer where a $4\pi M_{eff}$ is equal to 12.5 kOe has been measured.

Figures 4a and 4b show the typical MS-FMR angular dependence of the resonance field at different frequency for 20 and 50 nm thick films. The MS-FMR measurements show that the angular behavior of resonance fields is governed by the term of uniaxial anisotropy. The uniaxial anisotropy field, presented on figure 4c, shows a similar behavior as the coercive field and increases linearly with the CFA inverse thickness. This uniaxial anisotropy maybe due to the substrate vicinal structure induced by

the miscut, due to the enhancement of the CFA chemical order [14] or due to the growth-induced strain. However, the easy direction of the uniaxial anisotropy, determined with a 90° precision since it is referenced with a respect to substrate edges, changes with the thickness (Fig. 2e) and complicates the identification of its origin. Therefore, a completely satisfactory interpretation of the presence of H_u and its thickness dependency is still missing. Assuming that the dependence of the thickness is due to uniaxial surface anisotropy ($H_u = H_u^V + 4K_u^S/M_S$), the uniaxial surface anisotropy constant estimated from the linear fit of uniaxial anisotropy field is $K_u^S = 7 \times 10^{-3}$ erg/cm². For CFA films grown on Si substrates, the absence of the in-plane fourfold anisotropy, observed in the case of CFA thin films deposited on MgO [9,14], is in good agreement with the above-mentioned in-plane random orientation crystal growth of the CFA films grown on Si substrates. For all the samples, the obtained values of the magnetic parameters corresponding to the best fits of the MS-FMR measurements have been used to fit the MOKE measurements where a good agreement has been obtained.

V. CONCLUSION

Co₂FeAl films of various thicknesses (10 nm < d < 100 nm) were prepared by sputtering on a Si(001)

substrates and annealed at 600°C. They show an out-of-plane epitaxial growth with an in-plane polycrystalline structure. The out-plane lattice parameter increases with increasing CFA due to the enhancement the chemical order or the residual strain. MOKE hysteresis loops obtained with different field orientations revealed that the easy axis coercive field varies linearly with the inverse CFA thickness. The microstrip ferromagnetic resonance has been used to study the dynamic properties. The angular and the field dependences of the resonance field and frequency, respectively, have been analyzed through a model based on a magnetic energy density which, in addition to Zeeman, demagnetizing and exchange terms, is characterized by a uniaxial anisotropy to determine the most relevant parameters. The in-plane uniaxial anisotropy field, present in all the samples, increases linearly with the inverse CFA thickness. However, the uniaxial anisotropy easy axis direction changes with the thickness. The effective magnetization shows drastically linear increase with the inverse CFA thickness due to the enhancement of the CFA/MgO interface quality.

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