Effect of Film Thickness on the Structural and Magnetic Properties of Co$_2$FeAl$_{0.5}$Si$_{0.5}$ Heusler Alloy Thin Film

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Abstract. Co$_2$FeAl$_{0.5}$Si$_{0.5}$(CFAS) thin films of thickness, $t = 25$ nm, 50 nm and 75 nm have been investigated using ferromagnetic resonance (FMR) to determine the Gilbert damping constant, effective magnetization, $g$-factor, and anisotropy field. Structural characterization reveals that the film with thickness, $t = 50$ nm, has the highest B2 crystalline order, highest magnetization ($\sim$ 1304 Gauss), lowest inhomogeneous contribution to linewidth and lowest Gilbert damping constant ($\sim$ 0.002). The temperature variation of saturation magnetization, $M_S(T)$, is best described by the Bloch $T^{3/2}$ law of thermal demagnetization along with the thermal renormalization term in the spin-wave stiffness expression.

INTRODUCTION

In the wake of novel spintronic devices and applications, systems with 100% spin polarization have gained immense interest, both in the basic and applied research. Extensive studies have been carried out in order to understand the structural, magnetic properties and spin-polarization of Co-based Heusler alloys [1], which are theoretically predicted to be 100% spin polarized systems [2-4]. In conjunction with the spin polarization of a system, the damping parameter of a ferromagnetic material is one of the vital magnetic properties in order to realize high speed magnetization switching [5] and for spin-transfer-driven magnetic reversal [6]. Understanding the relaxation mechanisms in magnetic thin films can pave way towards spintronic advances. Ferromagnetic resonance (FMR) studies have been employed to explore the magnetic properties: effective magnetization, $g$-factor, effective magneto-crystalline anisotropy field and the nature of anisotropy through the FMR spectra [7-9]. FMR also provides an advantage over the conventional magnetometry in thin films characterization, where the substrate contribution (diamagnetic) to the sample (film) magnetization can be effectively tackled. In this work, the effect of film thickness on the magnetization, magnetic anisotropy and Gilbert damping in Co$_2$FeAl$_{0.5}$Si$_{0.5}$(CFAS) has been investigated using a broad-band FMR. Furthermore, an attempt to discern the mechanisms contributing to the temperature dependence of saturation magnetization, $M_S(T)$, has been made.

EXPERIMENTAL DETAILS

Co$_2$FeAl$_{0.5}$Si$_{0.5}$(CFAS) thin films of thickness, $t = 25$ nm, 50 nm and 75 nm, were deposited by ultrahigh vacuum dc magnetron sputtering on Si(100) substrates with SiO$_2$ buffer layer (300 nm), at the optimum substrate temperature of 500°C. CFAS thin films were deposited from a stoichiometric CFAS target (Co: 50; Fe: 25; Al: 12.5; Si: 12.5). Deposition rate of 0.04 nm/sec was achieved at an optimized power of 40W and 5 mTorr argon pressure. The deposition rate was optimized by measuring the thickness as a function of time using a surface profilometer. The structural characterization was performed by High-resolution Grazing angle X-ray diffraction (HR-GIXRD) (D8 discover Bruker X-ray diffractometer) with a Cu (K$_\alpha$) source, at a grazing angle of $\omega = 0.5^\circ$. The elemental analysis
and chemical composition of the as-deposited CFAS films were determined using an energy dispersive X-ray spectroscope (EDS), which is an accessory to the field emission scanning electron microscope (FESEM). Frequency dependent ferromagnetic resonance (FMR) measurement was performed using NanoOsc PhaseFMR instrument with coplanar waveguide (CPW) sample stage attached with a 15 kOe electromagnet. In this, the static magnetic field (H) is swept perpendicular to the applied microwave field (AC) along the in-plane (IP) direction of the thin film at fixed microwave frequencies ranging from 8 GHz to 39 GHz. Temperature dependent FMR over a temperature range 120 K to 300 K was measured by using BRUKER-A300 ESR spectrometer with an X-band frequency of 9.5 GHz.

RESULTS AND DISCUSSION

FIGURE 1. (a) GIXRD pattern of 25 nm, 50 nm and 75 nm CFAS thin films deposited on SiO\(_2\)/Si(100), (b) Field derivative of microwave power absorption, dP/dH, as a function of H for t = 50 nm at fixed frequencies ranging from 8 GHz to 39 GHz.

Figure 1(a) shows the GIXRD patterns for the CFAS films. Observation of the Bragg peaks, corresponding to the fundamental reflections (220), (400) and (422) confirms the growth of crystalline order in the system. The even superlattice reflection (222) in t = 25 nm and 50 nm, affirms that the CFAS films with thickness t = 25 nm and 50 nm has B2 crystalline structure [3, 4] with t = 50 nm having the highest B2 order. By contrast, in the t= 75 nm film, the (222) diffraction peak is missing, suggesting the existence of A2 order.

Figure 1(b) shows the field derivative of microwave power absorption, dP/dH, plotted against H for t = 50 nm at fixed frequencies ranging from 8 - 39 GHz, recorded in the IP configuration, i.e., the static magnetic field (H) is varied perpendicular to the applied microwave filed along the film plane at fixed microwave frequency. In order to obtain the resonance field, \(H_{res}\), and the line width, \(\Delta H\), from the FMR spectra, asymmetric Lorentzian function is employed which is given by [9, 10],

\[
\frac{dS_{12}}{dH} = a - b \cdot \frac{2\Delta H^2(H-H_{res})}{[(H-H_{res})^2+\Delta H^2]^2} - c \cdot \frac{\Delta H[(H-H_{res})^2-\Delta H^2]}{[(H-\Delta H^2)^2+\Delta H^2]^2}
\]

(1)

where \(\frac{dS_{12}}{dH} \propto \frac{dP}{dH}\), and c are the coefficients of the absorptive (symmetric) and dispersive (anti-symmetric) term in Eq.(1)

Figure 2(a) and 2(b) shows frequency dependence of \(H_{res}\) and \(\Delta H\) for t = 50 nm, obtained from the best fit using Eq.(1). The frequency dependence of \(H_{res}\) is well described by the Kittel resonance condition [11],

\[
\omega = \gamma \sqrt{(H_{res} + H_k) \times (H_{res} + H_k + 4\pi M_s)}
\]

(2)

where, \(\gamma = g\mu_B/\hbar\) is the gyromagnetic ratio, \(g\) is the Landé splitting factor, \(M_s\) is the saturation magnetization, \(H_k\) is the IP anisotropy field. \(M_s\) and \(H_k\) thus obtained from the best fit using Eq. (2) are plotted as a function of thickness in Fig.2(c). It can be seen that the film with t = 50 nm has the highest magnetization, \(M_s = 1304(1)\) Gauss and highest value of \(H_k \sim 36\) Oe confirming the increased crystalline order. Line width \(\Delta H\) as a function of
frequency is analysed with the Landau-Lifshitz-Gilbert (LLG) equation \[12, 13\] along with the extrinsic \(\Delta H_{inh}\) contribution,

\[
\Delta H = \Delta H_{inh} + \frac{2}{\sqrt{3}} \gamma 2\pi f
\]

where, \(\Delta H_{inh}\) contribution to linewidth arises from sample roughness, porosity etc., and \(\alpha\) is the intrinsic Gilbert damping constant in the LLG term. The variation of \(\alpha\) with the film thickness is shown in Fig.2(d). The film with thickness \(t = 50\) nm has the lowest \(\alpha (~ 0.002)\), in good agreement with the reported values [14].

**FIGURE 2.** (a) Frequency dependence of \(H_{res}\), (b) Frequency variation of \(\Delta H\) for \(t = 50\) nm, (c) \(H_k\) and \(M_{eff}\) as functions of thickness, (d) Variation of Gilbert damping constant with thickness.

**FIGURE 3.** (a) Temperature dependence of \(H_{res}\) in IP-easy axis (\(\varphi_H = 0^\circ\)) and IP-hard axis (\(\varphi_H = 90^\circ\)), (b) Temperature dependence of \(\Delta H\) in IP-easy axis (\(\varphi_H = 0^\circ\)) and IP-hard axis (\(\varphi_H = 90^\circ\)) for \(t = 50\) nm, (c) Temperature variation of \(H_k\) for \(t = 50\) nm, (d) Temperature variation of \(M_S\) (for \(t = 50\) nm) with fit using spin-wave expression (red continuous line).

Temperature dependence of \(H_{res}\) and \(\Delta H\) in IP-easy axis (\(\varphi_H = 0^\circ\)) and IP-hard axis (\(\varphi_H = 90^\circ\)) is shown in Fig. 3(a) and 3(b). \(H_{res}\) increases with temperature while \(\Delta H\) show a decreasing trend with increasing temperature. The relation, \(H_k(T) = \frac{[H_{res}(T, \varphi_H = 90^\circ) - H_{res}(T, \varphi_H = 0^\circ)]}{2}\), is used to determine the anisotropic field \(H_k\) and, in turn, obtain the saturation magnetization \(M_S\) from the resonance condition. Figures 3(c) and 3(d) show the
temperature variations of $H_s$ and $M_s(T)$, respectively. To determine the exchange stiffness, $M_s(T)$ is fitted using the spin wave expression [15]. The main observation is that the Bloch $T^{3/2}$ along with the thermal renormalization term of spin wave stiffness, $D(T) = D(0) \left[1-D_2 T^2 \right]$, best describes the $M_s(T)$.

SUMMARY AND CONCLUSION

In summary, Co$_2$FeAl$_{0.5}$Si$_{0.5}$ thin films with thickness 50 nm has the highest crystalline B2 order and highest magnetization. Lowest Gilbert damping constant is obtained at $t = 50$ nm with $\alpha = 0.002$. The linewidth analysis of FMR spectra confirms that the film with $t = 50$ nm has the least $\Delta H_{\text{inh}}$ contribution. The temperature dependence of $M_s(T)$ obeys the $T^{3/2}$ and the thermal renormalization term becomes important in describing the $M_s(T)$ behaviour.

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