Thin Solid Films 558 (2014) 349–355

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Effect of sputtering pressure and power on composition, surface roughness, microstructure and magnetic properties of as-deposited Co_2FeSi thin films

K. Srinivas *, M. Manivel Raja, D.V. Sridhara Rao, S.V. Kamat

Defence Metallurgical Research Laboratory, Hyderabad 500 058, India

ARTICLE INFO

Article history:
Received 6 September 2013
Received in revised form 22 January 2014
Accepted 12 February 2014
Available online 20 February 2014

Keywords:
Heusler alloy thin films
A2 structure
TEM
Magnetic properties
DC magnetron sputtering

ABSTRACT

The effect of sputtering pressure and power on the composition, surface roughness, structure, microstructure and magnetic properties of full Heusler type Co_2FeSi films deposited on silicon (001) substrates at room temperature using DC magnetron sputtering technique was studied. The composition of the film was found to be influenced by both sputtering pressure and power. The films deposited at lower pressures (0.667–2 Pa) and lower power (50–75 W), which result in lower deposition rates, have compositions comparable with that of the target. The surface roughness of the films was found to be significantly affected by a change in sputtering pressure but not by a variation in sputtering power. Irrespective of the sputtering pressure and power, all the as-deposited Co_2FeSi/Si(001) films exhibited A2 type disordered nanocrystalline structure. The grain size, however, was found to increase marginally with an increase in sputtering pressure and power. Analysis of magnetization curves of the films revealed that the films were soft ferromagnetic for all sputtering pressures and power. However, the coercivities, magnetization values and reversibility of zero field cooling and field cooling magnetization curves were found to depend critically on sputtering parameters.

1. Introduction

Recently, full Heusler type half-metallic ferromagnets (HMFs) have received a considerable attention because they are predicted to have 100% spin polarization at the Fermi energy level (E_F) [1]. These alloys thus have a significant potential for spintronic applications such as in tunnelling magneto resistance based (TMR) magnetic tunnel junction (MTJ) devices [2], spin injection/detection devices [3], spin filters [4] and nonvolatile magnetoresistive random access memory (MRAM) [5]. Among full Heusler alloys, Co_2FeSi has been found to be a promising candidate due to its high magnetic moment (~6 μ_B/f.u) and high Curie temperature (~1100 K) [6]. Co_2FeSi based alloys also have the greatest potential to exhibit half-metallicity and high TMR at room temperature [7]. Moreover, Co_2FeSi has a conductivity of the same order as non-magnetic semiconductors and low lattice mismatch of ~4% with Si (001). Thus if Co_2FeSi thin films possessing stoichiometric composition, smooth surfaces, ordered structure, high magnetic moment and half-metallic character can be grown on Si substrates, it can be used as an efficient spin polarized electrode facilitating an integration of spintronic devices into Si based very large-scale integrated circuits.

The first magnetic tunnel junction with Co_2FeSi thin films (with $L2_1$ type structure) was prepared by Inomata et al. [8] on MgO substrates with 52% TMR at room temperature. Daibou et al. [9,10] fabricated TMR junctions with Co_2FeSi (with A2 type structure) on thermally oxidized Si substrate using MgO as a barrier, which resulted in 90% TMR at ambient temperature after annealing at 598 K. It was also reported that it is not necessary to use a highly oriented epitaxial films as a ferromagnetic electrode [10–12] in TMR junctions. It is also well known that the spin polarization of HMFs is often affected by deposition conditions and chemical disorder [13]. Lee et al. [14] have also shown that the structural disorder affects weak-localization phenomena and canted spins in the ferromagnetic ground state and thereby leads to spin disorder scattering and negative high-field magneto resistance. This shows that the deposition conditions of the films play an important role.

It is well known that DC magnetron sputtering technique for thin film deposition has an advantage of higher productivity due to low deposition temperatures and relatively higher deposition rates compared to other deposition methods and hence is widely used as a mass production process. Previous studies on sputtered Co_2FeSi films [9,15–18] have shown that sputtering parameters have a large influence

* Corresponding author. Tel.: +91 40 24586783; fax: 91 40 24340884.
E-mail addresses: kuchanasrinivas@gmail.com (K. Srinivas), mraja@dmrl.drdo.in (M. Manivel Raja), dvsridhararao@gmail.com (D.V. Sridhara Rao), kamat@dmrl.drdo.in (S.V. Kamat).

http://dx.doi.org/10.1016/j.tsf.2014.02.052
0040-6090/

on the composition and structural quality of Co_2FeSi films deposited on MgO (001) substrates. In fact, it has been observed that non-stoichiometric films result even when stoichiometric targets are used during deposition by magnetron sputtering if the sputtering conditions including substrate temperature are not optimized [15]. It should be noted that although high deposition temperatures result in well ordered Heusler alloy films, high deposition temperatures are prohibited for the deposition of these films in most spintronic devices because of the requirement of smooth surfaces, sharp interfaces and minimized inter-diffusion between the films and substrates [19]. This suggests that the optimization of sputtering parameters is critical especially the most important parameters such as sputtering pressure and power. Although several investigators have studied the structural and magnetic properties of Co_2FeSi films grown by sputtering technique [9,15–18], systematic studies on the effect of sputtering power and sputtering pressure on the composition, surface roughness, structure and magnetic properties of Co_2FeSi thin films grown at room temperature on Si (001) substrate by DC sputtering have not yet been carried out. This kind of study is essential if Heusler alloys are to be used in spintronic devices in the future.

In light of the above, in the present investigation, the effect of sputtering pressure and power on composition, surface roughness, structure, microstructure and magnetic properties of Co_2FeSi thin films deposited on Si(001) substrates at room temperature was studied.

2. Experimental

Two groups of Co_2FeSi thin film samples were deposited by DC magnetron sputtering. In one group, the sputtering pressure was altered (0.667, 1.34, 2, 2.67 Pa) with other parameters (time, substrate to target distance) unchanged, while in the other group the sputtering power was varied (25, 75, 100 and 125 W) with other parameters unchanged. High purity argon (Ar) gas was used as a sputtering gas. For all the films, target-substrate distance and the sputtering time were kept constant at 102 mm and 30 min, respectively. Additionally, in the first set of films, the sputtering power was kept constant at 50 W and in the second set of films, the sputtering pressure was kept fixed at 1.34 Pa. All the films were deposited on single crystalline Si (001) substrates at room temperature using a DC magnetron sputtering system with a $Co_{49.95}Fe_{24.98}Si_{24.99}$ (3″ size) alloy target. The composition of the target was determined using inductive coupled plasma optical emission spectroscopic (ICP-OES) analysis and chemical analysis. From these results, it was found to be very close to the stoichiometric composition i.e. $Co_{50}Fe_{25}Si_{25}$. Prior to commencing sputtering, the base pressure of the chamber was maintained below 2.67×10^{-5} Pa for 2 h and pre-sputtering was done for 60 s to remove any impurities present on the target surface. The deposition was carried out on a deoxidized Si (001) substrates treated with standard wet chemical treatment. The thickness of the films was determined by measuring the step height by means of a stylus profilometer. Stylus profilometer was also used to determine the surface roughness of the films. The crystal structure was determined by X-ray diffraction (XRD) using a Phillips X'pert diffractometer (Cu K_α ($\lambda = 1.5406$ Å)) as well as electron diffraction patterns using a transmission electron microscopy (TEM). The cross-sectional images and surface morphology of the films were investigated using field emission scanning electron microscope (FESEM), TEM and Atomic Force Microscope (AFM). Composition was measured using energy dispersive spectroscope (EDS) attached to TEM and also cross-checked with those of films deposited on Al_2O_3 substrate by EDS attached to FESEM. Magnetic measurements such as magnetization versus applied field (M–H) plots at 4 K and 300 K and zero field cooling (ZFC) and field cooling (FC) measurements were undertaken with the help of super conducting quantum interference device (SQUID) (Model: MPMS SQUID VSM (Ever Cool)-SVSM072).

3. Results and discussion

3.1. Thickness, surface roughness and composition

The evaluated thickness and surface roughness values of all the as-deposited films with varying sputtering pressure and sputtering power are given in Table 1. It can be seen that the film thickness increases with an increase in both sputtering pressure as well as sputtering power in the ranges studied. The most important parameter during sputtering, as mentioned earlier, is the deposition rate which depends critically on sputtering pressure and power. In the present investigation, the deposition rate is found to increase from 6.5 to 9.7 nm/min with increasing sputtering pressure from 0.667 to 2.67 Pa. These observations are consistent with the fact that in the pressure range studied in this investigation, the sputtering yield increases with increasing pressure but the pressure is not high enough to reduce the mean free path of the target atoms due to collisions with the Ar ions. The deposition rate also increases from 4.5 to 12.5 nm/min with increasing sputtering power from 25 to 125 W. This is consistent with the fact that increasing sputtering power results in an increase in the kinetic energy of the target atoms. It is also clear that the enhancement of deposition rate with increasing sputtering power is more than that of increasing sputtering pressure in the ranges studied. This can be attributed to the fact that the higher kinetic energy of the target atoms results in higher surface diffusions once these atoms are adsorbed on the substrate surface. It is also observed that the surface roughness values increase with an increase in sputtering pressure but are not significantly affected by the change in sputtering power in the ranges studied.

The compositional details of all the thin films are also given in Table 1. With increasing sputtering pressure, the Si and Fe contents are found to decrease while Co content is found to increase. In case of the films deposited at different sputtering powers, Co and Fe contents are found to decrease while Si content is found to increase significantly. The films deposited at lower pressures (0.667–2 Pa) and lower power (50–75 W) which result in lower deposition rates have compositions comparable with that of the target. These results also indicate that sputtering power has more influence on the film composition than the sputtering pressure in the ranges studied.

3.2. Structure and microstructure

Fig. 1 shows the XRD patterns of the as-deposited Co_2FeSi films on Si(001) substrates with varying sputtering pressure (Fig. 1(a)) and sputtering power (Fig. 1(b)). It can be observed that in all cases the Co_2FeSi thin films are crystalline in the as-deposited condition and have a body centred cubic A2 type crystal structure (space group: *Im*3*m*). (110), (002) and (211) diffraction peaks are observed irrespective of the sputtering pressure or power. The intensities of (110) and (211) peaks decrease with increasing sputtering pressure but they do not change much with increasing sputtering power. The broader full width at half maxima of (110) and (002) peaks clearly indicates that average crystallite sizes are very small. The average crystallite sizes from X-ray diffraction patterns of the films were estimated using the Williamson–Hall method [20] and enumerated in Table 1.

In order to understand the effect of the sputtering parameters on the microstructure of the films, all samples were studied using TEM, FESEM and AFM. Figs. 2 and 3 show typical microstructures and corresponding electron diffraction images of Co_2FeSi thin films deposited at 0.667 Pa/50 W and 1.34 Pa/100 W, respectively, recorded by using TEM, FESEM and AFM. The FESEM images reveal that all the as-deposited films are continuous, but consist of grains approximately 10–30 nm in diameter (Table 1). The samples deposited at lower sputtering pressure and power exhibit smaller grains (~10–15 nm) and the grain size increases marginally with increasing sputtering