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Annealing induced low coercivity, nanocrystalline Co–Fe–Si thin films exhibiting inverse cosine angular variation

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ABSTRACT

Co–Fe–Si based films exhibit high magnetic moments and are highly sought after for applications like soft under layers in perpendicular recording media to magneto-electro-mechanical sensor applications. In this work the effect of annealing on structural, morphological and magnetic properties of Co–Fe–Si thin films was investigated. Compositional analysis using X-ray photoelectron spectroscopy and secondary ion mass spectroscopy revealed a native oxide surface layer consisting of oxides of Co, Fe and Si on the surface. The morphology of the as deposited films shows mound like structures conforming to the Volmer–Weber growth model. Nanocrystallisation of amorphous films upon annealing was observed by glancing angle X-ray diffraction and transmission electron microscopy. The evolution of magnetic properties with annealing is explained using the Herzer model. Vibrating sample magnetometry measurements carried out at various angles from 0° to 90° to the applied magnetic field were employed to study the angular variation of coercivity. The angular variation fits the modified Kondorsky model. Interestingly, the coercivity evolution with annealing deduced from magneto-optical Kerr effect studies indicates a reverse trend compared to magetisation observed in the bulk. This can be attributed to a domain wall pinning at native oxide layer on the surface of thin films. The evolution of surface magnetic properties is correlated with morphology evolution probed using atomic force microscopy. The morphology as well as the presence of the native oxide layer dictates the surface magnetic properties and this is corroborated by the apparent difference in the bulk and surface magnetic properties.

1. Introduction

Soft magnetic thin films are a hot topic of research due to their wide ranging applications in various fields such as magnetic recording, MEMS, sensors, etc., [1–3]. Soft magnetic properties are related to various factors such as alloy composition, nature of magnetic phase, crystal structure, crystal size and annealing conditions. In this context, Co–Fe based materials assume importance, owing to their high saturation magnetization and promising high frequency characteristics. The $Co_{70}Fe_{30}$ composition is thermodynamically stable in the bcc crystal structure [4]. This composition [4] is close to the maximum of spin polarization and possesses the maximum magnetic moment, as shown by the Slater Pauling curve [5]. Combination of these properties makes compositions near to $Co_{70}Fe_{30}$ suitable for various applications such as spin injection systems in spintronic devices [6].

Si can be added to Co–Fe alloys to facilitate amorphisation as well as to tune the magnetic properties. If the material can be tailored in to nanocrystalline thin films then they can be integrated in microelectronic devices. Co–Fe/Co–Fe–Si based metallic glasses are available commercially and these materials can be processed itnto nanocrystalline form by thermal annealing and they possess excellent soft magnetic properties, suitable for applications in transformer cores and magnetic shielding. Amorphous thin films of Fe–Ni/Fe–Ni–B which were subsequently processed into nanocrystalline form by annealing were recently reported [7–13]. However amorphous/nanocrystalline thin films of Co–Fe–Si have not been studied in detail or are seldom reported. Hence a detailed investigation of the nanocrystallization and change of magnetic properties of Co–Fe–Si thin films with thermal annealing was conducted.

Co–Fe thin films are usually prepared on different seed layers to reduce their coercivity. Thomson et al. reported coercivity of 16 Oe for Co–Fe films grown on Au/MgO seed layers [14]. Platt et al. reported coercivity of 12 Oe for Co–Fe films deposited on CoO. They showed that domain walls in the soft films have relatively large mobility in response to changing magnetic fields below the nominal H_c. They also attributed the observed low coercivity values

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to the averaging of the high anisotropy energy [15]. Vopsaroiua et al. reported the dependence of coercivity on grain size for 20 nm Co–Fe thin films prepared by sputtering. They observed a reduction in the coercivity from 120 Oe for samples with a mean grain size larger than 17 nm down to 12 Oe for a sample with a mean grain size of 7.2 nm [16]. Ji reported the growth and physical properties of epitaxial $Co_{70}Fe_{30}$ thin films on Si substrate with a TiN buffer layer. They also reported that the films prepared at 450 °C exhibit a biaxial stress up to 0.52%. The films were reported to have a small in-plane biaxial anisotropy, low coercivity of 23 Oe for film thickness greater than 30 nm [17]. In the above mentioned investigations, the magnetic properties were explained using the Random Anisotopy model (RAM) initially proposed by Alben et al. [18] and modified by Herzer [19]. According to Herzer, excellent soft magnetic properties can be realized, if the size of the individual magnetic domain is reduced to the exchange coupling length (*L*ex). The subsequent exchange coupling results in very low local anisotropies and demagnetization effect. Hence for a large number of exchange coupled nanocrystalline grains, the local anisotropy K_1 is small, resulting in lowering of coercivity. Thus, Co–Fe films can be prepared with very low coercivity by the deposition of an under layer or upper layer of an cobalt ultrathin cobalt oxide. Most of the earlier reports focus on the bulk magnetic property of films, probed using VSM. Hence, detailed investigations, comparing, the surface and bulk magnetic properties of Co–Fe–Si thin films are yet another motivation for this work.

There are several reports on the magnetization reversal mechanism in soft magnetic thin films [6,17]. The Stoner–Wolfarth model based on coherent rotation [20] and the Kondorsky model based on domain wall motion/unpinning [21] are the two important models used for explaining the angular variation of coercivity. Even though one expects structures with sizes below the single domain size to obey the coherent rotation model, size dependent behavior was also observed [22]. In most ferromagnetic materials magnetization reversal is affected by domain nucleation and growth. In the coherent rotation model one assumes that the magnetic vectors rotate collectively with the applied field before reaching saturation. In contrast, the Kondorsky model assumes that magnetization reversal is primarily affected by nucleation and growth of reverse domains or the strong pinning of domains at local defects and inhomogeneities and predicts a $1/\cos\theta$ dependence of coercivity, where θ is the angle between the easy axis and the applied magnetic field. The Herzer model predicts similar coercivity variations for magnetization reversal by coherent rotation and domain wall motion/unpinning models [19].

Even though the Kondorsky model was originally derived for explaining the angular variation of coercivity in hard magnetic materials [21], similar behavior has been observed in many soft magnetic systems [23,24]. Thomson et al. [25] reported that the magetisation reversal of large soft magnetic islands of Co–Pt takes place by nucleation of a 180° reverse domain, followed by the spread of a domain wall throughout the islands. Delalande et al. observed the Kondorsky type angular variation of reduced coercivity in soft magnetic Co–Pt systems with perpendicular anisotropy [26]. Streubel et al. modeled the angular variation of magetisation reversal in Fe–Ni caps by the modified Kondorsky relation [27]. Spiridis et al., based on magnetic studies conducted on Co thin films of various thicknesses, reported that as film thickness decreases, the magetisation reversal mechanism can change from coherent rotation to domain wall movement [28]. Liu et al. reported the Kondorsky type dependence in cobalt thin films [29]. The in plane easy axis coercivity variation with grain size in [15–17] Co–Fe thin films was explained using the Herzer model. However no systematic investigation regarding the easy to hard axis magnetization reversal of Co–Fe-Si thin films has been reported in the literature. Hence investigations on the angular variation of magnetization reversal in Co–Fe–Si thin films assume significance.

We report the deposition of magnetic thin films of Co–Fe–Si on glass and NaCl substrates and the evolution of their magnetic properties with thermal annealing. The films exhibit onset of nanocrystallisation and grain growth with annealing. Further the morphology shows a profound change with annealing which is reflected in change in surface magnetic properties investigated using the magneto-optical Kerr effect (MOKE). The Herzer model is invoked to explain the observed soft magnetic properties of ultra-thin magnetic films. The angular variation of coercivity from in plane to out of plane shows an inverse cosine relationship exhibiting a Kondorsky type variation. The magnetization reversal is primarily governed by the pinning of domains at local defects.

2. Experimental

Thin films were vacuum evaporated using tungsten filaments at a vacuum of 10^{-6} Torr on NaCl and chemically cleaned glass substrates. A composite target with a composition corresponding to $Co_{69}Fe_4Ni_1Mo_2B_{12}Si_{12}$ was used for evaporation. Samples deposited on NaCl were used for TEM analysis. The thicknesses of the deposited films were determined using a Dektac 6M Stylus Profiler. The thin film samples were annealed at 100, 300 and 400 °C for 1 h under a high vacuum of 10^{-6} Torr to avoid possible surface oxidation. GXRD measurements were carried out on the annealed and pristine samples using a Bruker D8 Discover diffractometer with monochromatic Cu Kα X-rays at a grazing incidence angle of 0.5° and wavelength 1.5414 Å. XPS study of the films deposited on float glass substrates was performed with an Omicron Nanotechnology XPS system with monochromatic Al Kα radiation ($h\upsilon = 1486.6$ eV) of source voltage 15 kV and emission current of 20 mA. All scans were carried out at an ultrahigh vacuum of 1.5×10^{-10} Torr. The obtained XPS spectra were deconvoluted and quantified using Casa XPS program (Casa Software Ltd., UK), in which the background was simulated using the Shirley function and the peaks were fitted using a Gaussian Lorentzian function. The spectrum recorded was corrected using the binding energy of adventitious carbon at 284.6 eV and the accuracy of the measured binding energy values is estimated to be ± 0.2 eV. The elemental composition of the sample is extracted from the wide scan, while the individual element peaks were analyzed to obtain the chemical composition. As charging effects are unavoidable in the XPS study of thin films deposited on non-conducting samples, charge compensation was performed by electron gun flooding. The nanoscale imaging was performed using atomic force microscopy (AFM) (Digital Instruments Nanoscope V) in the tapping mode, using ultrahigh resolution cantilevers made of tungsten having radius of less than 1 nm and force constant of 46 N/m. Room temperature magnetization measurements were carried out using VSM (DMS 1660 VSM) with field varying from −10 to +10 kOe. The angular variation of magnetization is recorded by measuring the magnetization with the sample positioned at different angles with respect to the applied field. When the field is along the plane of film the angle is 0° and out of plane the angle is 90°. The surface magnetic properties were probed using a MOKE setup, operated using a red laser with 6328 Å wavelength from a He–Ne laser source. The loops were recorded with a magnetic field applied along the in plane direction.

3. Results

3.1. Composition analysis

The average thickness of the films was found to be 54 nm using the stylus profilometer.