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Research articles

Training effect of the exchange bias in sputter deposited Fe_3O_4 thin films with varying thickness

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ABSTRACT

The training effect property of the exchange bias in the reactively sputtered polycrystalline Fe_3O_4 thin films of varying thicknesses in the range 25–200 nm are studied. Structural studies by X-ray diffraction, X-ray photoelectron spectroscopy and selected area electron diffraction confirm the formation of single phase Fe_3O_4. The scanning electron spectroscopy images show that the grains are uniformly distributed. All the samples show clear and consistent exchange bias training behaviour due to the dynamics of the spins at the interface of the ferrimagnetic core and the spin glass-like surface of the grains. The analysis of the training effect data of the exchange bias field H_E measured at 2 K by using three different models show that the model based on the relaxation of the frozen and rotatable spin components at the interface gives the best description for all the samples. From this model, it is found that the reversible interface spins relax around 7 times faster than the frozen interface spins at 2 K for all the samples and that their relative relaxation rates are independent of the sample thickness. This constancy show that the relative relaxation rates of the interfacial frozen and rotatable spin components is a material dependent property. The frozen component of the interfacial spins of each sample is found to be dominated at the initial stage of the training. A direct equivalence between the H_E and remanence asymmetry M_E is observed. Above the spin freezing temperature, the training effect measurements at 75 K show that the H_E decreases sharply with successive field cycling as compared to the measurements made at 2 K and the H_E vanishes after first few cycles.

1. Introduction

Exchange bias is the phenomenon of coupling which usually develops when materials have ferromagnetic (FM) – antiferromagnetic (AFM) interface and this gives rise to unidirectional magnetic anisotropy. This in general is identified as the shift of the hysteresis loop along the field axis when the system is field cooled (FC) through the Neel temperature (T_N) of the AFM. Exchange bias effect was first discovered by Meiklejohn and Bean [1] in a compact of nanoparticles having core-shell structure where the fine Co particles coated with a shell of antiferromagnetic CoO. Since then the exchange bias has been observed in variety of systems include FM/AFM thin film heterostructures with a diverse combination of materials [2,3], FM or ferrimagnetic nanoparticles embedded in an AFM matrix [4], materials having cluster glass or spin glass phases [5,6]. Over the years the exchange bias effect has been extensively studied both for the fundamental understanding as well as because of its immense technological applications in spintronic devices such as magnetic recording read heads [7], magnetic random access memories [8,9], giant magnetoresistive sensors [10] and spin valves [11]. The exchange biased nanoparticles can stabilize well against thermal fluctuation and hence become advantageous for biomedical applications [4]. For device applications a robust and durable exchange bias is required. The stability of the exchange bias can be evaluated by an aging phenomenon known as the training effect. The training effect refers to the gradual and monotonous degradation of the exchange bias shift along the field axis upon cycling the system through consecutive hysteresis loops at a fixed temperature [2,12]. In general, the training effect arises due to irreversible changes in the magnetic microstructure of the AFM, as its spin structure rearranges with each magnetization reversal of the FM layer. Despite of the intensive experimental and theoretical research works, the underlying microscopic mechanism responsible for the exchange bias as well as the training effect are still debated.

In our earlier report [13], we have studied the origin of the exchange bias in polycrystalline films of Fe_3O_4 which is a ferrimagnetic half-metallic material having high spin polarization with Curie temperature T_C = 860 K and hence it is among the potential

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materials for spintronic applications. The samples we studied are pure Fe_3O_4 which did not have an AFM layer of a separate material. The exchange bias of the Fe_3O_4 thin films were also investigated by some authors [14–17]. The existence of exchange bias, especially in epitaxial Fe_3O_4 films is often attributed to the presence of the antiphase boundaries [14,15]. Unlike the epitaxial Fe_3O_4 films, our study [13] revealed that the exchange bias effect in our polycrystalline films is originating mainly because of the existence of a significant exchange coupling between the core spins and the spin glass-like surface spins of the Fe_3O_4 grains. This is due to the fact that each grain of these polycrystalline films can be modeled as core-shell like structure. The core is the inner portion of the grain having regular atomic arrangement and the spins are well-aligned ferrimagnetically. The shell is formed by the atoms located at the surface of the grains. Due to the bond order deficiencies of these surface atoms, uncompensated surface spins are formed. These spins can display spin glass-like behaviour due to various magnetic interactions experienced at the surface. We have observed a significant increase in H_E at temperatures below the spin freezing temperature (T_f) which is below ~50 K. This is due to the freezing of the spin glass-like surface spins in the direction of the cooling field below T_f. Such exchange bias behaviour in polycrystalline Fe_3O_4 films arising from the spin glass-like grain boundaries is also reported by some authors [15–17]. As a continuation of our research work, in the present article we report the training effect study on the polycrystalline Fe_3O_4 films of varying thicknesses. The samples were successfully prepared at ambient temperature by reactive dc magnetron sputtering. The present study gives further insights into the phenomenon of exchange bias in our system having ferrimagnetic-spin glass like layer interfaces by analyzing the data using different models.

2. Experimental details

The Fe_3O_4 thin films of thickness (t) ranging from 25 to 200 nm were deposited at ambient temperature on to Si(1 1 1) substrates by reactive dc magnetron sputtering using a Fe target. The sputtering was carried out in a mixture of argon and oxygen gas ambience with an argon gas partial pressure $P_{Ar} = 4 \times 10^{-3}$ mbar. The oxygen gas partial pressure (Po_2) was maintained precisely in order to obtain the single phase Fe_3O_4 films of well defined stoichiometry. The thicknesses of the films was measured using a Bruker DektakXT profilometer and was further confirmed through X-ray reflectivity (XRR) measurements performed by using a Rigaku high resolution X-ray diffractometer (HRXRD) with Cu-$K_{\alpha 1}$ radiation. Structural studies of the samples were performed using the same HRXRD system in a grazing incidence geometry and by X-ray photoelectron spectroscope (XPS) of Thermo VG Scientific-MULTILAB 2000 with Al-K_α radiation. Microstructural analyses were performed by High resolution transmission electron microscope (HRTEM) of JEOL JEM 2100F with an accelerating voltage of 200 kV and high resolution Field emission gun scanning electron microscope (FEGSEM) of JEOL JSM 7600F. Magnetization measurements were carried out by a SQUID-VSM of Quantum design, Inc.

3. Results and discussion

3.1. Structural and microstructural characterization

Grazing incidence X-ray diffraction (GIXRD) measurements on the Fe_3O_4 thin films were carried out by keeping a glancing angle of $\omega = 0.5°$. As a representative, the data for the 100 nm thick Fe_3O_4 film is shown in Fig. 1(a). The positions of the diffraction peaks and their relative intensities seen in this figure are consistent with that of the powder XRD data of the cubic inverse spinel crystal structure of Fe_3O_4. This indicates that the films are polycrystalline with no preferred orientation. The grain sizes (D) of the samples were calculated from the (3 1 1) peak by using Scherrer formula [18]. The calculated values of D are 6.3, 8.5, 13.6 and 21.5 nm, respectively, for the 25, 50, 100 and 200 nm thick films. These D values are found to be directly proportional to the thickness of the films.

The γ-Fe_2O_3 (maghemite) phase also has a cubic inverse spinel crystal structure similar to Fe_3O_4 phase and hence these two phases cannot be distinguished by the XRD data alone. We have performed X-ray photoelectron spectroscopy (XPS) on our samples

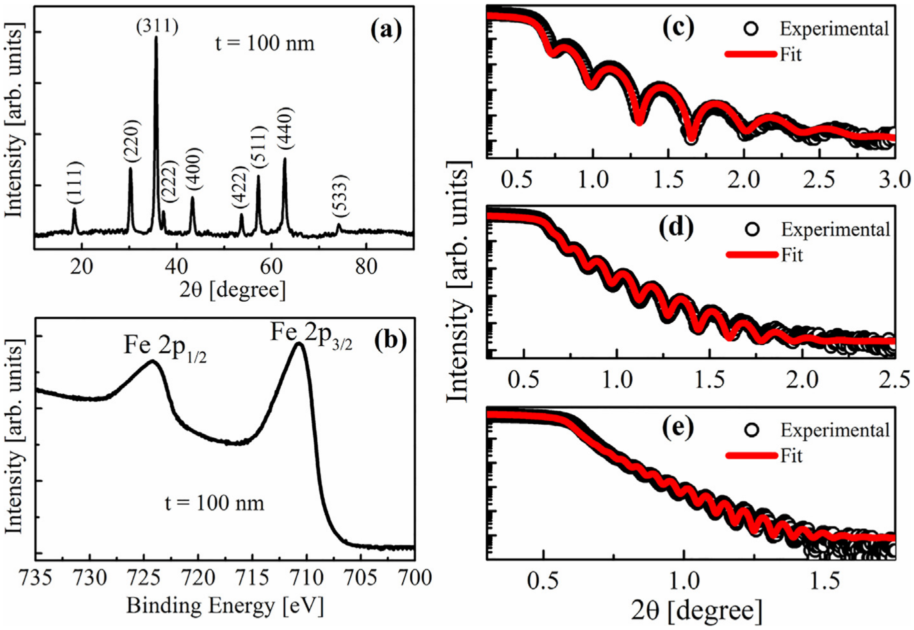

Fig. 1. (a) Grazing incidence X-ray diffraction data of the 100 nm thick Fe_3O_4 film. (b) X-ray photoelectron spectroscopy core level (Fe 2p) spectrum of the same 100 nm thick film. (c)–(e) X-ray reflectivity patterns for the Fe_3O_4 films having different thicknesses. The nominal thicknesses of the films are (c) 25 nm (d) 50 nm and (e) 100 nm. Circles are the experimental data and the solid lines represent the fits.