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Modification of magnetic anisotropy induced by swift heavy ion irradiation in cobalt ferrite thin films

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ABSTRACT

The present study demonstrates the modification of magnetic anisotropy in cobalt ferrite ($CoFe_2O_4$) thin films induced by swift heavy ion irradiations of 200 MeV Ag-ion beams. The study reveals that both magnetizations and coercive field are sensitive to Ag-ions irradiation and to the fluences. The magnetic anisotropy enhanced at low fluence of Ag-ions due to domain wall pinning at defect sites created by ion bombardment and at high fluence, this magnetic anisotropy ceases and changes to isotropic behavior which is explained based on the significant structural and morphological changes. An X-ray absorption and x-ray magnetic circular dichroism studies confirms the inverse spinel structure of these compounds.

1. Introduction

The directional dependence of magnetic properties or anisotropy of magnetic materials are of great interest to the researchers, for the light they throw on basic magnetic phenomena, and to technologist, who may exploit them in the design for specific applications [1]. Depending on the type of application, material with high, medium or low magnetic anisotropy will be required [2]. Magnetic thin films with high coercivity and anisotropies are intensively investigated today, in order to satisfy the demand of increasing magnetic storage density [1–6]. With large anisotropy, the superparamagnetic limit can be pushed down, and a stable magnetization can be promoted in ultra small nanosized magnetic bit, which are needed in advanced media for ultrahigh density recording [7]. Magnetic anisotropy can result from several sources. It can be due to crystal structure i.e. crystallographic texture, the internal stress or strain as well as from the particular shape or arrangement of particles and any one of them may become predominant under special circumstances [1]. In reduced symmetry system like thin films, besides intrinsic crystal anisotropy, extrinsic anisotropy like shape, stress anisotropies are drastically enhanced; competes with each other and may lead to reorientation or modification of anisotropy depending on the predominant source of anisotropy [7]. Therefore, understanding the sources of anisotropies and their engineering is crucial for the technological use. Among the various magnetic materials, $CoFe_2O_4$ (cobalt ferrite) thin films has attracted much attention due to their unusual magnetic properties such as high coercivity, moderate saturation magnetization (M_s), large magneto-crystalline anisotropy and magnetostriction [6–13]. Compared to other materials which possess high coercivity, $CoFe_2O_4$ have the advantage of being low cost and excellent chemical stability, and its large magnetostriction can promote the stress anisotropy to prevail in low dimensional structures such as thin film [4,7]. $CoFe_2O_4$ has an inverse spinel structure, symbolized as $[Fe^{3+}]_A[Co^{2+}Fe^{3+}]_BO_4$, in which one-eighth of the tetrahedral sites (A-sites) are occupied by Fe^{3+} ions and one-half of the octahedral sites (B-sites) are occupied by Co^{2+} and Fe^{3+} ions. It is reported that a small fraction of Co^{2+} ion is located on the tetrahedral sites depending on the thermal history of the sample [14–16]. Cations (Co^{2+} and Fe^{3+} ions) on the A and B-sites are coupled antiferromagnetically via superexchange interaction whereas cations are ferromagnetically coupled within A or B-sites. $CoFe_2O_4$ owing to a spin-orbit stabilized ground state (with unquenched orbital momentum $l_z = \pm 1$) caused by a trigonal crystal field on the Co^{2+} octahedral cations results in a large magnetocrystalline anisotropy [17,18]. Therefore, cationic arrangement and strain effects can dramatically change the magnetic properties of $CoFe_2O_4$ thin films [11,19].

The magnetic anisotropy can be tuned by varying thickness of

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film, lattice mismatch between the film and the substrate and processing temperatures [3–5,7,11,12,20–23]. Lisfi et al. have reported reorientation of magnetic anisotropy in epitaxial $CoFe_2O_4$ thin films upon varying the film thickness. The driving force of such a phenomenon is due to be the lattice strain [7]. Gao et al. show that the magnetic anisotropy of epitaxial $CoFe_2O_4$ films grown on $SrTiO_3$ substrates can be reoriented by inserting a thin $SrRuO_3$ buffer layer. The $CoFe_2O_4$ films have a uniaxial anisotropy with the easy axis perpendicular to the film plane but with the insertion of $SrRuO_3$ buffer layer results in the switching of the easy axis into the in-plane orientation. That is associated with a tensile and a compressive strain for the films without and with buffer layer [21]. The switching of magnetic anisotropy from out-of-plane to in-plane associated with a tensile and a compressive strain have also been demonstrated by Dhakal et al. on $CoFe_2O_4$ films grown on MgO and $SrTiO_3$ substrates [22]. Raghunathan et al. studied the variation of perpendicular anisotropy and coercivity in $CoFe_2O_4$ films grown on SiO_2/Si (100) with substrate temperature and reported that the perpendicular magnetic anisotropy decreased with decreasing substrate temperature due to the thermal expansion mismatch between the film and the substrate [23].

Swift Heavy Ion (SHI) irradiation is a unique and effectual tool which is known to generate controlled defects, structural disorder and can modify strain in thin films [24,25]. SHI during their passage through material result in excitation and ionization of the atoms by inelastic collisions (called as electronic energy loss, S_e) or displace them by elastic collisions (nuclear energy loss, S_n). These are known to result in production of defects in the target [26]. The shape of the defects introduced by SHI irradiation depends on the S_e of the impinging ions. The defects morphology changes from a cluster of spherical defects to cylindrical defects above a threshold value of electronic energy loss (S_{eth}) in magnetic insulators [27,28]. In complex system like ferrites; where cations reside in two different sublattices, the defects can disturb the arrangement of cations which may result in significant effects on magnetic properties including the magnetic anisotropy. The advantage of SHI irradiation is that the properties can be locally and controllably changed depending upon the electronic energy loss and fluence values in the target materials [26]. The present study focuses on the modification of magnetic anisotropy in $CoFe_2O_4$ thin films by SHI irradiation using 200 MeV Ag^{+17} ions at different fluences.

2. Experimental details

Thin films of $CoFe_2O_4$ of thickness about 150 nm have been grown onto SiO_2/Si (100) substrates by Pulsed Laser Deposition (PLD) method using 284 nm KrF excimer laser with energy 188 mJ at substrate temperature 500 °C and oxygen gas pressure of 20 mTorr. The base pressure of the chamber was 6×10^{-6} Torr. The laser repetition rate and the distance between the target-substrate were maintained at 10 Hz, 5 cm respectively. After deposition, it was cooled to the room temperature under same oxygen pressure. The films were irradiated with 200 MeV Ag^{+17} ions at the fluences 5×10^{11}, 1×10^{12} and 5×10^{12} ions/cm^2 using 15UD Pelletron at Inter – University Accelerator Centre (IUAC), New Delhi. The direction of incidence of Ag beams is 0° relative to the sample normal. The beam size is about 2 mm diameter and is uniformly scanned using magnetic scanners to an area of 15×15 mm^2 and the samples of size 5×5 mm^2 were kept within this area, therefore the angular spread at the edges of the samples is neglected since the energy of incident ion is very high and roaster scanning process is fast. The structural characterizations were carried out with X-ray diffraction (XRD) θ–2θ scans and Raman Spectroscopy and cross-section Transmission Electron Microscope (XTEM). Surface morphology and magnetic domain structure were determined by Atomic Force Microscopy (AFM) and Magnetic Force Microscopy (MFM) respectively. Magnetic hysteresis loops were measured at room temperature using a Vibrating Sample Magnetometer (VSM). To understand their electronic structures, X-ray absorption spectroscopy (XAS) and X-ray magnetic circular dichroism (XMCD) experiments were performed at dragon beamline (BL11A1) at NSRRC, Hsinchu, Taiwan. The measurements were performed in an ultrahigh-vacuum (UHV $\approx 10^{-10}$ mbar) chamber and magnetic field upto ± 1 Tesla is applied along the easy axis using circularly polarized beam. XAS spectra were recorded in the total-electron-yield (TEY) mode on measuring the sample drain current and normalized to the incoming flux (I_0) as measured on an Au-mesh at the entrance of the experimental chamber. The degree of circular polarization was 80%.

3. Results and discussions

Fig. 1 shows the XRD pattern of the pristine and irradiated $CoFe_2O_4$ thin films and inset showing the shifting of peak position of (400) plane. The pristine film has diffraction peaks which correspond to crystalline cubic spinel structure (JCPDS File no. 22-1086) but the peak intensities are quite different which indicate preferred orientation. The intensity ratios of (222)/(311), (333)/(311) and (400)/(311) are 23%, 49% and 71% respectively, which is larger than the corresponding values of 6%, 26% and 20% for the conventional polycrystalline target. Since ratio is largest for (400)/(311), the preferred orientation is along (100). The peaks for pristine thin film are observed to be shifted towards the lower angles as shown in the inset of Fig. 1 which indicate the presence of strain. The intensity of all the peaks decreases with increase in fluences value. Partial amorphization of the film created by ion irradiation leads to the decrease of peak intensity. 200 MeV of Ag ion corresponds to $S_e \sim 21$ KeV/nm was larger than or equal to the threshold (1–20 KeV/nm) of formation of homogeneous cylindrical amorphous tracks or columnar amorphization [29–32]. The creation of columnar amorphization is understood based on thermal spike model [33]. In this model, most of the energy of the incident ions is transferred to the lattice by electron-phonon interaction. This results in a rise of local lattice temperature far above the

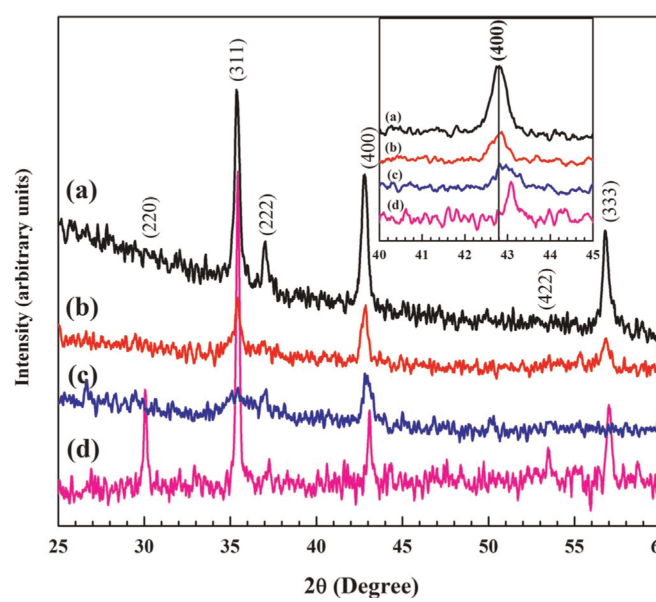

Fig. 1. XRD pattern of the pristine and irradiated $CoFe_2O_4$ thin films and the Inset shows the enlarged view of (400) (a) pristine (b) 5×10^{11} ions/cm^2 (c) 5×10^{12} ions/cm^2 and (d) target.