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Influence of 150 MeV Ni^{11+} swift heavy ion irradiation on $CuFe_2O_4$ thin films prepared by radio frequency magnetron sputtering: Modification on structure and surface morphology

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ABSTRACT

The bulk copper ferrite nanomaterials are synthesized by co-precipitation technique. The vibrating sample magnetometer measurement for bulk $CuFe_2O_4$ reveals its unsaturated superparamagnetic behavior. The crystallites of the synthesized nanomaterial are in cubic form having an average size of ~100 Å and are used as target to prepare thin films of various thicknesses (600, 900 and 1100 nm) by radio frequency magnetron sputtering technique. X-ray peaks arise only when films are annealed at 1200 °C and also they are found to be in tetragonal structure. The magnetic characteristics of 900 nm unirradiated $CuFe_2O_4$ thin film exhibit superparamagnetic behavior and have an unsaturated magnetic moment at high field. Magnetic force microscopy images of unirradiated $CuFe_2O_4$ thin films confirm the soft nature of the magnetic materials. The 150 MeV Ni^{11+} swift heavy ion irradiation on these thin films at the fluence of 1×10^{14} ions/cm^2 modifies the polycrystalline nature due to electron–phonon coupling. Atomic force microscopy image shows that the swift heavy ion irradiation induces agglomeration of particles in 600 and 900 nm thin films and increases rms surface roughness from 33.43 to 39.65 and 69.7 to 102.46 nm respectively. However, in 1100 nm film, holes are created and channel-like structures are observed along with a decrease in the rms surface roughness from 75.12 to 24.46 nm. Magnetic force microscopy images of 900 nm irradiated thin film explain the formation of domains on irradiation and are also supported by the ferromagnetic hysteresis observed using vibrating sample magnetometer.

1. Introduction

The developments of ferrite materials have received much attention in the field of magnetic memory device fabrication and biological industry recently. The square and rectangular planar hysteresis ferrites are used in the fabrication of magnetic memory devices [1], while superparamagnetic nanoparticles have gained much interest in the field of biomedical applications [2]. Copper ferrite is generally in cubic structure [3] but transforms to tetragonal symmetry when slowly cooled from higher temperature to room temperature [4,5]. There are different methods such as solid state reaction, co-precipitation technique, sol–gel technique and self combustion technique to synthesize powder form of ferrite materials with improved properties for specific applications [6–10]. Different methods of fabrication of ferrite thin films and evaluation of their properties are reported [11–13]. However, solid state reaction method is the mostly used technique to synthesize the target material to prepare thin films by radio frequency (RF) sputtering technique [14,15]. Literature survey reveals that very few attempts have been made to use the ferrite material prepared by co-precipitation method as target to deposit thin films by RF sputtering technique [16]. Among the irradiation properties of thin films, modifications in physical parameters are much needed one. In swift heavy ion (SHI) irradiation, the incident ion losses its energy in two ways:(1) S_n— nuclear energy loss $(-(dE/dx)_n)$ and (2) S_e— electronic energy loss $(-(dE/dx)_e)$ [17,18]. In the nuclear energy loss, the energy is lost by elastic collisions of incident ions, which are dominant at low energies (<1 MeV/amu). In electronic energy loss process the dominant energy loss at high energies is due to inelastic collisions of ions with the atoms. At low energy irradiation, the ions get embedded and modify the material nature by the collision cascade produced by the impinging ions. In SHI irradiation, modifications occur in thin films due to the electronic excitation. In such a situation, the elastic collision effects causing collision cascade is avoided and the effect of the embedded ion does not arise [19]. It is well established that electronic stopping power and nuclear energy play a vital role in defect creation mechanism. SHI penetrates deep into the materials, produces a long and narrow disordered zone and sometimes creates perceptible path way (holes) along its trajectory [20,21].

In this paper, we report the influence of swift heavy ion irradiation on thin films of $CuFe_2O_4$ prepared by RF magnetron sputtering

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technique. The formation of magnetic domain, modifications in rms roughness and change in crystallinity by Ni^{11+} ion beam irradiation (150 MeV) on thin films are reported herein.

2. Experimental methods

2.1. Thin film preparation

The copper ferrite bulk material was prepared by standard co-precipitation technique. In order to obtain the desired compositions, stoichiometric amounts of 0.5 M $Cu(NO_3)_2.3H_2O$ and 1 M $Fe(NO_3)_3.9H_2O$ were dissolved in 250 ml of double distilled water with constant stirring. The neutralization was carried out with 4 M sodium hydroxide solution. The reaction temperature was kept at 85 °C for 1 h. The precipitate was thoroughly washed with distilled water until the pH reaches 7. The product was dried in an oven at 100 °C for overnight to remove the water contents. The sample was grinded into fine powder and then annealed at 250 °C for two and half hours and then cooled to room temperature with the same rate as that of heating. The X-ray diffraction (XRD) studies were carried out to confirm the spinel ferrite phase structure formation in the material and to test the quality of the basic material used for the investigation. The stoichiometric composition was verified by energy dispersive X-ray (EDAX) analysis. The copper ferrite disks of 50 mm in diameter and about 5 mm thickness were prepared, sintered at 750 °C for 2 h and used as a target. Thin copper ferrite films of different thickness were deposited on 1 mm thick quartz plates by RF magnetron sputtering technique using Hind Hivac system.

The target was placed about 6 cm away from the substrate. The base pressure was maintained at 6.67×10^{-4} Pa using diffusion pump before introducing the premixed Ar and O_2 sputtering gas into the chamber. To avoid any contamination on the target surface, to make the system stable and to reach optimum condition, the target was pre-sputtered for 15 min under 200 W RF power before deposition. The substrate temperature was monitored by a thermo-coupler placed near the substrate. The power and substrate temperatures were maintained at 200 W and 200 °C respectively. The schematic representation of $CuFe_2O_4$ thin film preparation using RF sputtering technique is shown in Fig. 1. The deposition was carried out for three different thicknesses 600, 900 and 1110 nm by varying the coating time 10, 20 and 60 min respectively. Thickness of the thin films was measured using surface roughness tester model surf test SJ 301 with a resolution of 0.01 μm.

2.2. Irradiation experiment

The irradiation experiments were performed on thin films using 150 MeV Ni^{11+} ions at the fluence of 1×10^{14} ions/cm^2 under high vacuum using 15 UD pelletron at Inter University Accelerator Centre, New Delhi. The selected zone area for beam scanning was 1 cm × 1 cm and the beam current used was 5 pnA. From the stopping range of ions in matter (SRIM) calculations, the range of the projected ions for 150 MeV Ni^{11+} is 8.65 μm. Since the films of different thicknesses 600, 900 and 1100 nm were subjected for irradiation, the ions are expected to pass through the film material without implantation of ions [22].

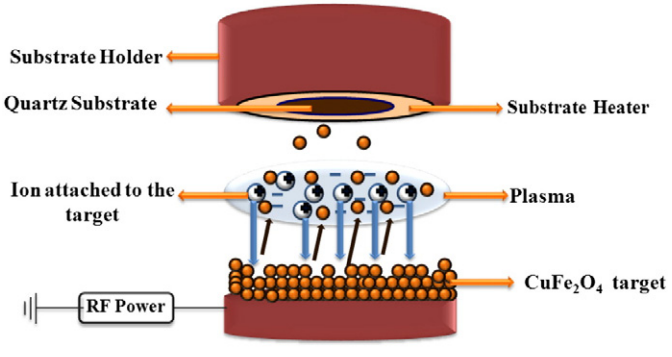

Fig. 1. Schematic representation of $CuFe_2O_4$ thin film preparation using RF sputtering technique.

2.3. Characterization

The XRD of bulk material and thin films was recorded with PANalytical X'Pert PRO X-ray diffractometer operated at 40 kV, 30 mA using Cu K_α radiation with a wavelength of 1.54060 Å using X'Celerator detector. The diffraction pattern was collected in steps of $2\theta = 0.05°$ and the counting time was 10 s per step. The composition of bulk materials was measured by JEOL JSM-5610 LV with INCA EDS instrument with an accelerating voltage of 20 kV at high vacuum mode. The semi quantification elemental analyses to identify the weight percentage of major and minor elements present in the samples were done using OXFORD INCA energy dispersive X-ray spectrometer. Atomic force microscopy (AFM) and magnetic force microscopy (MFM) images were taken in tapping mode at room temperature using NanoScope IIIa SPM. The noise floor for the instrument is $= \leq 0.05$ nm, which is the highest resolution possible for vertical measurement. The rms value of the AFM baseline noise is 0.01 nm. Each measurement was done at minimum two different regions to check the repeatability of the rms surface roughness. MFM measurements were carried out at constant lift height (50 nm) before and after irradiation without any applied magnetic field. An antimony doped silicon tip coated with magnetic layer of Co/Cr was used and scanned over the area 5 μm × 5 μm. The cantilever stiffness was 1–5 N/m. The mass magnetization of bulk material was measured using vibrating sample magnetometer (VSM) technique (Digital Measurement Systems — Model EV 9) in the applied field ranging from + 20 A/m to − 20 A/m. Similarly, the volume magnetization for thin films was carried out using 140 A/m VSM, Quantum Design, PPMS-VSM, in the field range + 40 A/m to − 40 A/m.

3. Results

3.1. XRD and EDAX analyses

The XRD pattern of bulk $CuFe_2O_4$ material prepared by co-precipitation method is shown in Fig. 2. The XRD pattern reveals the existence of copper ferrite spinel phase characteristic peak (311) along with (111), (220), (222), (400), (422), (511) and (440) X-ray peaks. It is well agreed with the standard JCPDS value (Card No. 77-0010). The measured d-space values are listed in Table 1. It is discernible from Fig. 2 that the copper ferrite has spinel face centered cubic structure. The cell

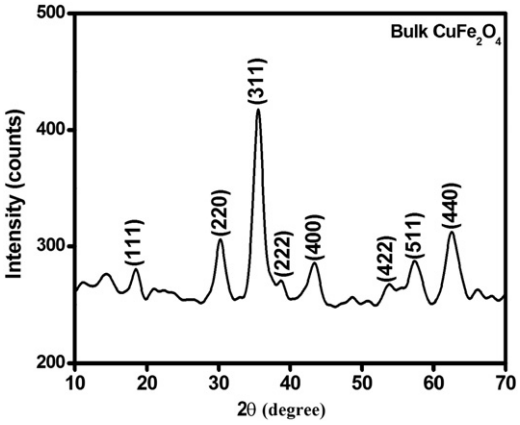

Fig. 2. X-ray diffraction pattern of bulk $CuFe_2O_4$.