Nuclear Instruments and Methods in Physics Research B 286 (2012) 277–281

Contents lists available at SciVerse ScienceDirect

Nuclear Instruments and Methods in Physics Research B

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

Structural and magnetic properties studies on swift heavy ion (SHI) irradiated Fe_3O_4 thin films

Jianrong Sun [a,*], Zhiguang Wang [a], Yuyu Wang [a], Yabin Zhu [a], Lilong Pang [a], Tielong Shen [a], Fashen Li [b]

[a] Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, China
[b] Key Laboratory for Magnetism and Magnetic Materials of the Ministry of Education, Lanzhou University, Lanzhou 730000, China

ARTICLE INFO

Article history:
Received 18 August 2011
Received in revised form 24 December 2011
Available online 10 January 2012

Keywords:
SHI irradiation
CEMS
MFM
Magnetic properties

ABSTRACT

Polycrystalline Fe_3O_4 thin films are synthesized at low temperature by electroless plating, and the effects of irradiation by Kr^{26+} ions at energy of 2.03 GeV on the magnetic properties of the Fe_3O_4 thin films are investigated by macroscopic magnetization measurements (vibrating sample magnetometer, VSM; superconducting quantum interference device, SQUID) and microscopic distribution of the magnetic moments (conversion electron mössbauer spectroscopy, CEMS) as well as magnetic domain structure (magnetic force microscope, MFM) analyses. The initial crystallographic structure of the Fe_3O_4 remains unaffected after swift heavy ion (SHI) irradiation at low damage levels, but VSM, CEMS and MFM results indicate that SHI irradiation is an effectual tool to not only modify the macro-magnetic properties (saturation magnetization, coercivity, etc.), but also modify the micro-magnetic properties (magnetic anisotropy, distribution of the magnetic moments, magnetic domain structure, etc.) significantly. And all modifications of these properties could be interpreted very well by the effects related to the stress and defects (the production, accumulation and free) induced by SHI irradiation.

1. Introduction

In recent years, as electronic devices have become much miniaturized with their high-performances advancing, the study of electronic devices under much higher-signal frequencies has become a trend. Metal alloys, as potential candidates, have limited applications because of their high conductivity and serious eddy currency loss at high frequencies. On the contrary, ferrites, owing to their large resistivity, low power loss, and high permeability, have become very important in high frequency soft magnetic applications pertinent to micro-inductors and micro-transformers [1]. Magnetite (Fe_3O_4) is this kind of typical soft magnetic ferrite material, with the properties of high saturation magnetization, large resistivity, high permeability, good mechanical hardness, and chemical stability, and has been widely used [2,3]. Moreover, Fe_3O_4 can be used as catalyst and in tunneling magnetoresistance (*TMR*) and giant magnetoresistive (*GMR*) devices [4–6]. In fact, Fe_3O_4 is highly attractive for spintronics applications because of its high spin polarization (ca. 100%) at the Fermi level and high Curie temperature (T_c) of 858 K, which is crucial for thermal stability in device applications [7–9].

However, the soft magnetic properties of polycrystalline Fe_3O_4 films were not that good and the expected significant *MR* effect was not observed, especially at room temperature [10]. Therefore, control and modification of magnetic properties are highly essential to obtain devices for magnetic and spintronics applications with improved performance characteristics. As is well known, swift heavy ion (SHI) irradiation is a unique and effectual tool to produce controlled defects, structural disorder, stress, and phase transformations in thin films and then to modify the physical properties of materials [11–13]. Conventional energetic ions lose energy in materials by inelastic and elastic collisions, and these energy losses are termed as electronic (S_e) and nuclear (S_n) stopping powers, respectively. But when the thickness of the targeted medium is sufficiently smaller than the range of the projectile ion, the energy deposition is mainly due to the S_e. In addition, the S_e process is more dominant in the case of SHI irradiation and the large value of energy transferred induces an unusual density of defects, stress and heat in the samples.

There have been reports on the modifications of magnetic properties in spinel ferrites after irradiation [14–16]. However, knowledge on the micro-magnetic properties' (distribution of the magnetic moments, magnetic anisotropy, magnetic domain structure, etc.) modification induced by SHI irradiation is still quite limited. So in the present work, we report on changes of the structural and micro-magnetic properties in Kr-ion irradiated Fe_3O_4 thin films.

* Corresponding author. Tel.: +86 9314969647; fax: +86 9314969330.
E-mail address: sunjr@impcas.ac.cn (J. Sun).

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doi:10.1016/j.nimb.2011.12.066

2. Experiment and discussion

Polycrystalline Fe_3O_4 thin films with thickness of 1 μm were synthesized on glass substrates by electroless plating in aqueous solution at 90 °C [17]. Then, the SHI irradiation experiment was performed on the materials research terminal of the HIRFL-SSC (IMP, Lanzhou). The parameters of irradiation experimental condition were given in Table 1. In experiment, Fe_3O_4 ferrite thin films were irradiated at RT with 2.03 GeV Kr^{26+} ions for fluence range from 5.0×10^{11} to 1.0×10^{13} ions/cm^2. These regimes produced four levels of radiation damage evaluated by SRIM 2008 in average 0.00005 displacement per atom (dpa), 0.0001 dpa, 0.0005 dpa and 0.001 dpa correspondingly. In other words, to understand the SHI irradiation performance of the Fe_3O_4 ferrite thin films at various and low levels of dpa, structural and magnetic properties studies have been carried out. The S_e, S_n and range of ion (R_p) have been calculated by using SRIM 2008 simulation program and are found to be 9.44 keV/nm, 4.60 eV/nm and 152.59 μm, respectively. From these values, it is clear that S_e is three orders of magnitude larger than S_n, and R_p is much larger than the film thickness, so the energy deposition is mainly due to the S_e. The following measurements were carried out for the deposited and irradiated films. The crystallographic structure and the surface morphology of the films were analyzed by X-ray diffraction with Cu Kα radiation (XRD, Philips X'pert, Holland) and scanning electron microscopy (SEM, Hitachi S4800, Japan), respectively. Composition of the films was investigated by using energy dispersive X-ray analysis (EDS, Genesis XM, USA) and X-ray photoelectron spectroscopy (XPS, PerkinElmer PHI-5702, USA). Conversion electron Mössbauer spectra were measured with a spectrometer (CEMS, Wissel, Germany). The γ-ray source of CEMS was a 25 mCi ^{57}Co in Pd. Vibrating sample magnetometer (VSM, Lakeshore 7304, USA) and superconducting quantum interference device (SQUID, Quantum Design MPMS XL-7, USA) were used to measure the magnetic properties of the ferrite films at different temperatures. A nanoscope multimode scanning probes microscope (SPM, Veeco Instruments NanoScope IIIa, USA) in the atomic force microscope (AFM) and magnetic force microscope (MFM) modes were used to determine surface roughness and magnetic domain structure of Fe_3O_4 thin films.

Fig. 1 shows XRD patterns of the films irradiated with different fluences. The results of XRD indicate that the initial crystallographic structure of the Fe_3O_4 films remains unaffected after Kr-ion irradiation, and all pristine and irradiated Fe_3O_4 films have a single spinel phase structure with polycrystalline columnar grains growing perpendicularly to the substrates. In consideration of the different variety of peak width and peak heights with different irradiation fluences, the value of the lattice constant (a) of the pristine and irradiated Fe_3O_4 films can be obtained with the equation $a = d_{hkl} \cdot \sqrt{h^2 + k^2 + l^2}$. It is also noted that the lattice constant is strongly affected by SHI irradiation with different fluences. Table. 2 shows the dependence of the lattice constant on the Kr-ion fluence for Fe_3O_4 thin films. The estimated lattice constants of all irradiated samples from their XRD patterns are systematically larger than the pristine sample's value of 8.411 Å, and the main reason is due to the stress effects induced by SHI irradiation. In other words, the changes in lattice constant mean that the stress of the films is different with different irradiation fluences. And the production, accumulation and relaxation of the stress induced by SHI irradiation can explain the changes in lattice constant very well.

Table 1
The parameters of irradiation experimental condition. Mean electronic stopping (S_e) and nuclear stopping (S_n) powers and mean projected ranges (R_p) obtained with the SRIM-2008 code for 1.0 μm-thick materials.

Material	Ion	Energy (GeV)	S_e (keV/nm)	S_n (keV/nm)	R_p (μm)	Fluence (ions/cm^2)
Polycrystalline Fe_3O_4	^{84}Kr	2.03	9.44	0.0046	152.59	5.0×10^{11} 1.0×10^{12} 5.0×10^{12} 1.0×10^{13}

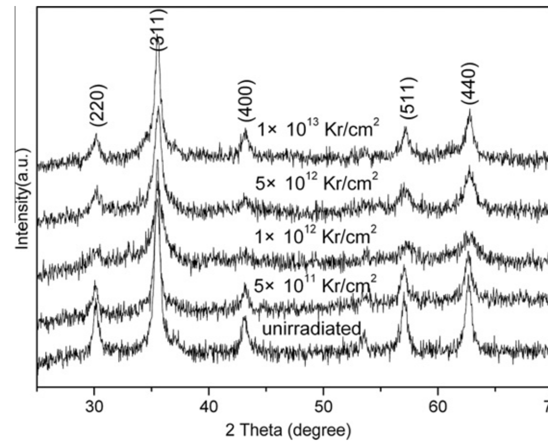

Fig. 1. XRD patterns of the Fe_3O_4 ferrite thin films during irradiation with 2.03 GeV Kr^{26+} ions at room temperature.

Table 2
The lattice constant of the Fe_3O_4 thin films with different irradiation fluences.

Fluence (ions/cm^2)	0	5.0×10^{11}	1.0×10^{12}	5.0×10^{12}	1.0×10^{13}
Lattice constant a (Å)	8.411	8.418	8.432	8.426	8.420

Fig. 2 shows the typical magnetic hysteresis loops of the pristine and irradiated Fe_3O_4 films. It is found that both saturation magnetization (M_s) and coercive force (H_c) of the Fe_3O_4 films are sensitive to Kr-ion irradiation and they exhibit different behaviors depending on the fluence ranges (Fig. 3). In fact, the energy transfer induced by SHI irradiation is a very complicated process. Notably, because the temperature of our films did not increase significantly, the effects of SHI irradiation on Fe_3O_4 films (production mechanism and nature of defects, atomic motion, relaxation, heat annealing effect, thermal spike effect, coulomb explosion, etc.) which are essentially due to the electronic stopping mechanism should be called electronic-excitation effects in general. And the modifications of magnetic

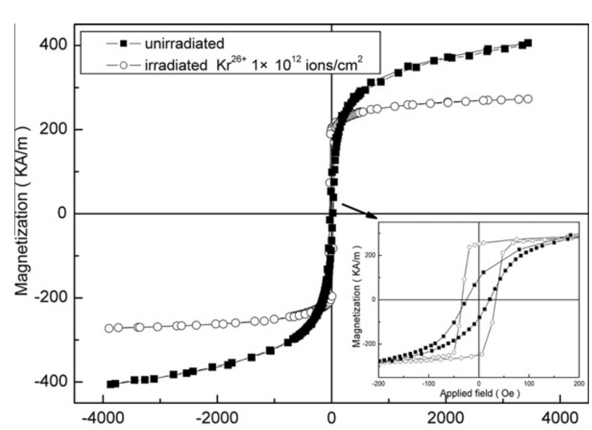

Fig. 2. Hysteresis loops at RT for the pristine and irradiated Fe_3O_4 films.