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Modifications in magnetic anisotropy of M—type strontium hexaferrite crystals by swift heavy ion irradiation.

Balwinder Kaur^a, Monita Bhat^a, F. Licci^b, Ravi Kumar^c, S.D. Kulkarni^d, P.A. Joy^d, K.K. Bamzai^a, P.N. Kotru^{a,*}

^aCrystal Growth and Materials Research Laboratory, Department of Physics and Electronics, University of Jammu, Jammu 180006, India

^bInstituto MASPEC – CNR, Via Chiavari 18/A, 43100 Parma, Italy

^cNuclear Science Centre, New Delhi 110067, India

^dPhysical Chemistry Division, National Chemical Laboratory, Pune 411008, India

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Abstract

Using vibrating sample magnetometery (VSM) 50 MeV Li^{3+} ion irradiation effects on magnetic properties of single crystals of $\text{SrGa}_x \text{In}_y \text{Fe}_{12-(x+y)} \text{O}_{19}$ (where x=0,5,7,9; y=0,0.8,1.3,1.0), are reported. The substitution of Ga and In in strontium hexaferrite crystals decreases the value of magnetization sharply, which is attributed to shifting of collinear magnetic order to a non-collinear one. Reduction of magnetization is also explained to be as a result of the occupation of the crystallographic sites of Fe^{3+} by Ga^{3+} and In^{3+} . The Li^{3+} ion irradiation decreases the value of magnetization, irrespective of whether the crystals are Ga–In substituted crystals of $\text{SrFe}_{12}\text{O}_{19}$. The result is interpreted in terms of the occurrence of a paramagnetic doublet in crystals replacing magnetic sextuplet as a result of irradiation. Substitution of Ga–In in Strontium hexaferrite decreases the value of anisotropy constant. Irradiation with Li^{3+} ions increases the values of anisotropy field for both substituted as well as unsubstituted crystals. Substitution with Ga–In also decreases the Curie temperature (T_c) but the irradiation with T_c ions does not affect the curie temperature of either Ga–In substituted or pure T_c crystals.

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1. Introduction

Hexagonal ferrites, the prototype of which is represented by MeFe₁₂O₁₉ (Me = Ba, Sr or Pb) belongs to a class of ferrimagnetic oxides. They have interesting applications not only as microwave devices but also as traditional permanent magnets, potential magneto-optical devices, high-density magnetic recording media (nanoscale) and compacts (submicron powders) for high-frequency devices [1,2]. Several attempts to improve the magnetic properties of these ferrites was done by substituting Fe³⁺ ions with some non-magnetic ions (i.e., Ga, In, Sc and Al) which may

result into materials that are paramagnetic at room temperature [3,4]. These materials may act as a suitable substrate for epitaxial deposition of magnetic hexaferrite films. These substitutions may lead to drastic variation in the magnetic structures of hexaferrites.

The intrinsic magnetic properties (i.e., saturation magnetization, anisotropy field) of these hexaferrites are strongly dependent on M-type phase [5]. The M-structure can be symbolically described as RSR*S*, where R is a hexagonal block containing Sr^{2+} ions, S is a spinel block, and the symbol * means that the corresponding block has been turned 180° around the hexagonal c-axis. In M-type ferrite, 12 Fe^{3+} ions per unit formula are distributed over five different sites in two blocks R (hexagonal) and S (spinel): three octahedral sites (12k, 2a and $4f_2$), one

^{*}Corresponding author. Tel.: +91 191 2450939; fax: +91 191 2453079. E-mail address: pn_kotru@yahoo.com (P.N. Kotru).

tetrahedral (4f₁) site and one pseudo-hexahedral site (2b) [5,6]. The 2b site is named so because the cation is displaced from the symmetry plane of the coordination trigonal bipyramid and plays an important role in determining the magnetic anisotropy of the M-type ferrites. The magnetic structure model as suggested by Gorter [7] is ferrimagnetic with five different sublattices, three (12k, 2a, 2b) parallel and two antiparallel (4f₁ and 4f₂) which are coupled by superexchange interactions through oxygen ions [5]. Table 1 (a) shows Fe³⁺ ions distributed among five different sublattices along c-axis [8]. The cell parameters for both unsubstituted and Ga–In substituted strontium hexaferrite as reported in the literature [4,9,10] is given in Table 1 (b).

M-type hexaferrites are well-known ferrimagnets with strong uniaxial anisotropy lying along the c-axis. This large value of magnetocrystalline anisotropy has its main origin in Fe³⁺ ions lying in 2b site that has a large asymmetry because a Fe³⁺ ion is surrounded by five oxygen ions [11]. Asti and Rinaldi [12] stressed that contribution to the anisotropy should be considered from all the five sites. This high anisotropy responsible for high Curie temperature and magnetic moment has made these hexaferrites good candidates in a number of applications. One of the important applications of these materials is in devices operating at higher microwave and millimeter wave frequencies. The high uniaxial anisotropy provides an internal magnetic field that suppresses the magnitude of the external applied field required for resonance at mm-wave frequencies [13]. This kind of devices requires high quality single crystals but the main drawback is strong uniaxial anisotropy that leads to low permeability and high resonant frequency. These drawbacks can be removed if one is able to decrease the anisotropy field or even modify the anisotropy from uniaxial to planar. The effect of substitution on magnetic properties is interesting for physics and applications. This high value of anisotropy can be reduced by substituting the Fe³⁺ ions by other

cations or mixture of cations (i.e., Ga, In, Sc, Al, Co, Ti, etc.). The substitution of Co–Ti [14,15], Co–Ru [16], Co–Ir [17–21] ions in M-type hexaferrites modify the anisotropy from uniaxial to planar. Extensive literature is available on the magnetic properties of the substituted hexaferrites [6,11,15,22–37]. Investigations [6,8,34–37] carried on substituted M-type hexaferrites show that non-collinear structures in the form of blocks and helical appears not only in the dispersed state but also in the bulk state. The selective occupation by non-magnetic ions i.e., In, Ga, Al, Sc at different crystallographic sites is responsible for altering exchange interactions between Fe³⁺ ions.

The irradiation-induced defects by swift heavy ions (SHI) have been extensively studied. It is reported [38] that corresponding to a large density of ionization and electronic excitation, amorphous latent tracks appeared in a material above a threshold of the electronic stopping power $S_e = (dE/dx)e$. It is quite interesting to understand the damage structures and modifications produced by swift heavy ion irradiation in magnetic oxides and ferrites [39–43]. Superexchange interactions between the magnetic ions are sensitive to any change in the crystallographic positions, which may result, into change in the magnetic properties in magnetic insulators like Y₃Fe₅O₁₂, BaFe₁₂O₁₉ or SrFe₁₂O₁₉. Mossbauer spectroscopy and vibrating sample magnetometry are two very convenient methods to study the ion irradiation effects. SHI irradiation-induced modification in magnetic materials have been studied extensively [10,39,42,44–50]. In magnetic materials like $Y_3Fe_5O_{12}$, BaFe₁₂O₁₉ and NiFe₂O₄, magnetic properties are very sensitive to irradiation-induced disorders which may result in a decrease of saturation magnetization [51-54]. Mossbauer spectroscopy has confirmed [55] that a paramagnetic phase is induced as a result of irradiation. It is also reported [49] that BaFe₁₂O₁₉ appears to be more sensitive to highenergy heavy ion irradiation than SrFe₁₂O₁₉.

Effect of SHI irradiation on mechanical characteristics of substituted and unsubstituted hexaferrites have already

Table 1

(a) Five iron sites, their spin direction, type, point symmetry, number of Fe ions per formula, and block situation in SrFe₁₂O₁₉

Sublattice	Type	Point symmetry	$Ions/SrFe_{12}O_{19}$	Spin $(5 \mu_B)$	Block
12k	Octahedral	m	6	Up ↑	S-R
$4f_1$	Tetrahedral	3m	2	Down ↓	S
$4f_2$	Octahedral	3m	2	Down ↓	R
2a	Octahedral	$\overline{3}$ m	1	Up ↑	S
2b	Five Fold	<u>6</u> m2	1	Up ↑	R

(b) Data show number of Sample	of Fe atoms substituted, their cell parameters corres Number of Fe atoms substituted		oonding to different compositions Cell parameter 'a' (Å)	Cell parameter 'c' (Å)
	(Ga)	(In)		
SrFe ₁₂ O ₁₉	0	0	5.833	23.05
$SrGa_5In_{0.8}Fe_{6.2}O_{19}$	5	0.8	5.884	22.93
$SrGa_7In_{1.3}Fe_{3.7}O_{19}$	7	1.3	5.888	22.88
$SrGa_9In_1Fe_2O_{19}$	9	1.0	5.924	22.98

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