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Modification of magnetic anisotropy in metallic glasses using high-energy ion beam irradiation

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Abstract. Heavy ion irradiation in the electronic stopping power region induces macroscopic dimensional change in metallic glasses and introduces magnetic anisotropy in some magnetic materials. The present work is on the irradiation study of ferromagnetic metallic glasses, where both dimensional change and modification of magnetic anisotropy are expected. Magnetic anisotropy was measured using Mössbauer spectroscopy of virgin and irradiated $Fe_{40}Ni_{40}B_{20}$ and $Fe_{40}Ni_{38}Mo_4B_{18}$ metallic glass ribbons. 90 MeV ^{127}I beam was used for the irradiations. Irradiation doses were 5×10^{13} and 7.5×10^{13} ions/cm^2. The relative intensity ratios D_{23} of the second and third lines of the Mössbauer spectra were measured to determine the magnetic anisotropy. The virgin samples of both the materials display in-plane magnetic anisotropy, i.e., the spins are oriented parallel to the ribbon plane. Irradiation is found to cause reduction in magnetic anisotropy. Near-complete randomization of magnetic moments is observed at high irradiation doses. Correlation is found between the residual stresses introduced by ion irradiation and the change in magnetic anisotropy.

Keywords. Magnetic anisotropy; metallic glasses; heavy ion irradiation; Mössbauer spectroscopy.

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1. Introduction

Energetic ions passing through any material deposit their energy by in-elastic and elastic collisions. Conventionally, these energy losses are termed as electronic (S_e) and nuclear (S_n) losses. At energies greater than 0.1 MeV per nucleon, the electronic energy loss, S_e, is predominant. The electronic energy losses are known to cause latent tracks in insulators [1] and recently have shown many effects on metallic materials such as, amorphization [2], phase transformation [3], annealing [4], damage creation [5] etc., when the S_e value crosses a certain threshold (S_{eth}). S_{eth} varies from 1 to 8 keV/Å, depending on the material. For metallic glasses the electronic energy loss is known to induce irreversible anisotropic change in the dimensions of a metallic glass ribbon [6]. Macroscopic growth of the dimensions perpendicular to the ion beam direction and shrinkage in the dimensions parallel to it are observed [6]. However, the total volume is conserved [6]. Recently, the electronic energy loss was shown to modify magnetic materials [7]. In some magnetic materials, the electronic energy deposition resulted in the change of magnetic anisotropy, because of the introduction of stresses [7,8]. The present work is on the irradiation study of ferromagnetic metallic glasses, where both dimensional change and modification of magnetic anisotropy are expected.

It is known that high-energy heavy ion irradiation induces strong local distortions and stresses throughout the irradiated region, which are radial and isotropic around the spherical defects [7]. The stresses induced by latent tracks in the surrounding material are able to rotate hyperfine magnetic field via magnetostriction coefficient, λ_s, in ferromagnetic materials [7]. After high-energy irradiation, garnet $Y_3Fe_5O_{12}$ and spinel $ZnFe_2O_4$ materials have shown magnetic field aligned parallel to the ion beam direction, i.e., compressive stress is induced which is in agreement with the negative value of λ_s [8], whereas after irradiation magnetite Fe_3O_4 shows magnetic field aligned perpendicular to the beam direction. It is known that annealing of metallic glasses can release the internal stresses and cause randomization of magnetic moments. In the case of a glassy ferromagnet $Fe_{40}Ni_{40}P_{14}B_6$ it has been found that annealing above 545 K causes randomization of magnetization axis [9]. In amorphous $Fe_{82}B_{12}Si_6$, upon annealing above 700 K, the rapid disappearance of perpendicular anisotropy was observed as a result of crystallization and weakening of compressive stresses [10]. For two ferrimagnetic oxides $Y_3Fe_5O_{12}$ and $BaFe_{12}O_{19}$, the enhancement of the paramagnetic phase is seen after the high-energy ion irradiation, when energy deposited to the electronic system (S_e) crosses a certain threshold [11]. It has also been observed that for higher value of S_e, anisotropy is observed in the bulk region beyond the range of high-energy ions [11]. Sorescu and Knobbe [12,13] have studied $Fe_{40}Ni_{38}Mo_4B_{18}$ by exposing it isochronally to pulsed-excimer-laser beam. It has been found that, at moderate values of repetition rate and pulse energy, controlled magnetic anisotropy could be induced in the higher magnetostriction samples without the onset of crystallization, whereas, random orientation of magnetic moment was obtained in the lower magnetostriction sample [12].

In the present work, we study the magnetic anisotropy induced by high-energy ion irradiation on $Fe_{40}Ni_{40}B_{20}$ and $Fe_{40}Ni_{38}Mo_4B_{18}$ metallic glasses and correlate it to the residual stresses introduced during dimensional change in metallic glasses. To the author's knowledge, no experiments are yet carried out to correlate the effects of dimensional change in metallic glass with the modification of magnetic anisotropy, both induced by high-energy ion irradiation.

2. Experimental

Commercially available $Fe_{40}Ni_{40}B_{20}$ and $Fe_{40}Ni_{38}Mo_4B_{18}$ metallic glass ribbons were cut into 1×1 cm^2 pieces. The samples were irradiated using 90 MeV ^{127}I beam to doses 5×10^{13} atoms/cm^2 and 7.5×10^{13} atoms/cm^2 in the Pelletron Accelerator at NSC, New Delhi. Thickness of the sample was about 25 μm. The range of the 90 MeV ^{127}I beam in this metallic glass is 7.2 μm. Thus, the ions lose their energy by elastic as well as in-elastic processes in the sample. Figure 1 shows that the electronic energy loss S_e and the nuclear energy loss S_n at various depths are produced in $Fe_{40}Ni_{40}B_{20}$ by 90 MeV ^{127}I beam. It can be seen that the predominant energy loss mechanism is via the electronic loss S_e. Mössbauer spectra were recorded for the samples before and after irradiation. A constant accelerator Mössbauer spectrometer was used in the standard transmission mode to record

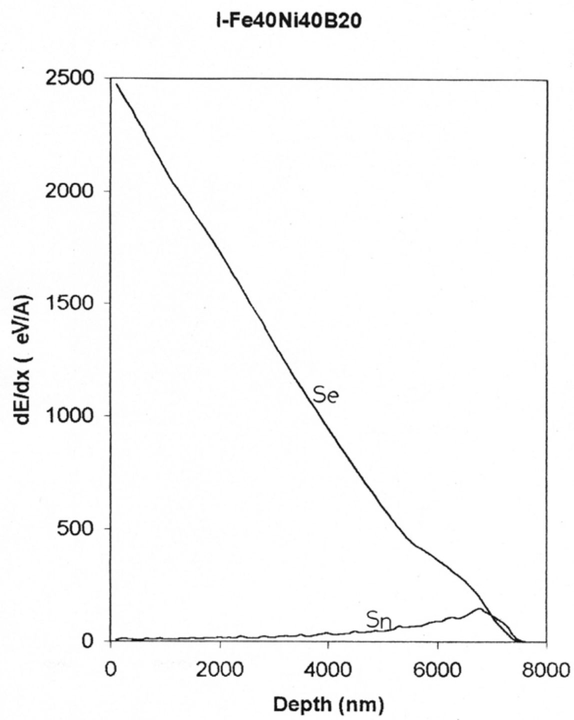

Figure 1. Electronic energy loss (S_e) and nuclear energy loss (S_n) of 90 MeV ^{127}I beam in $Fe_{40}Ni_{40}B_{20}$ metallic glass calculated using TRIM.

the spectra at room temperature. The source used was ^{57}Co in Rh matrix. The samples were placed perpendicular to the γ-ray direction. Recorded spectra were fitted using the NORMOS computer code developed by Brand [14]. The fittings used restricted $D_{13} = 3$, where D_{13} is the ratio of the intensities of the lines one and three in the magnetic sextuplet. The ratio D_{23}, which is the ratio of the intensities of the second and third lines, was used as a free parameter in the fitting programme. From D_{23} and D_{13}, D_{21} can also be calculated, where D_{21} is the ratio of the intensities of the second and first lines. D_{21} and D_{23} give the information about the magnetic anisotropy. D_{23} is given by

$$D_{23} = \frac{4\ \sin^2\langle\theta\rangle}{1+\cos^2\langle\theta\rangle} \tag{1}$$

where $\langle\theta\rangle$ is the average angle between the incident γ-ray and the average magnetic moment. The canting angle $\langle\theta'\rangle$ is defined as the angle between the average magnetic moment and the ribbon plane which is given by [15,16]

$$\langle\theta'\rangle = 90° - \arcsin\left[\frac{(3/2)D_{21}}{1+(3/4)D_{21}}\right]^{1/2}. \tag{2}$$

When the spins are aligned parallel to the ribbon plane (i.e., for ‘in-plane anisotropy’), we have $\langle\theta\rangle = 90°$, $\langle\theta'\rangle = 0°$, $D_{21} = 1.33$ and $D_{23} = 4$. When the spins are aligned perpendicular

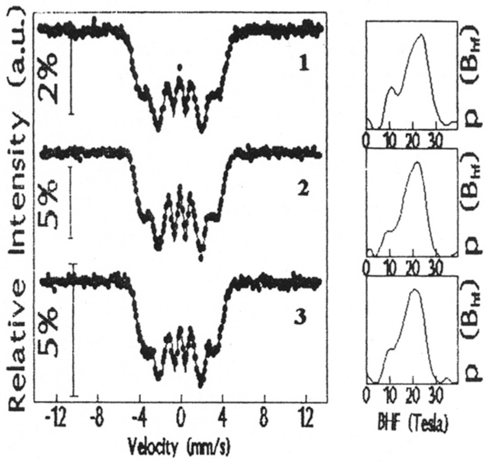

Figure 2. Mössbauer spectra of $Fe_{40}Ni_{38}Mo_4B_{18}$ metallic glass sample for (1) virgin, (2) irradiated with dose 5×10^{13} atoms/cm^2, and (3) irradiated with dose 7.5×10^{13} atoms/cm^2.

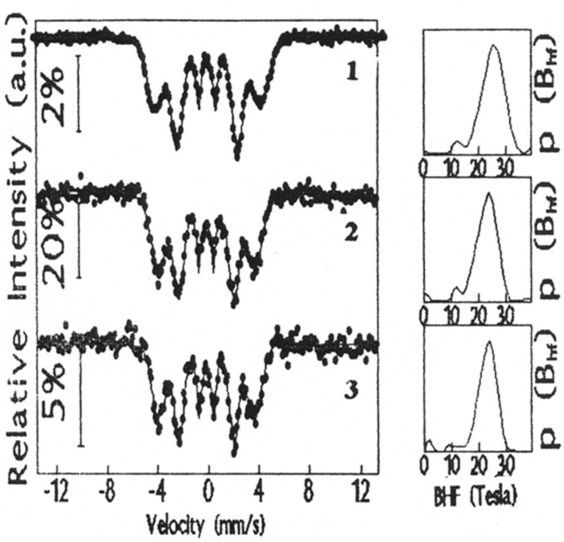

Figure 3. Mössbauer spectra of $Fe_{40}Ni_{40}B_{20}$ metallic glass sample for (1) virgin, (2) irradiated with dose 5×10^{13} atoms/cm^2, and (3) irradiated with dose 7.5×10^{13} atoms/cm^2.

to the ribbon plane (i.e., for 'perpendicular anisotropy'), we have $\langle\theta\rangle=0°$, $\langle\theta'\rangle=90°$, $D_{21}=0$ and $D_{23}=0$. When the magnetic moments are randomly aligned, we have $\langle\theta\rangle = 54.73°$, $\langle\theta'\rangle = 35.27°$, $D_{21} = 0.66$ and $D_{23} = 2$.

3. Results and discussion

Mössbauer spectra for both $Fe_{40}Ni_{40}B_{20}$ and $Fe_{40}Ni_{38}Mo_4B_{18}$ metallic glasses, for virgin as well as the samples irradiated with two different doses, are given in figures 2 and 3 along with the hyperfine field distributions found in the samples. The Mössbauer parameters for the same are listed in tables 1 and 2. It can be seen that the hyperfine field distributions do not vary significantly after irradiation except for a small shift at the lower values.

For $Fe_{40}Ni_{38}Mo_4B_{18}$ virgin sample, D_{23} is 2.35 and canting angle is 30.6°, indicating the presence of a small in-plane anisotropy in the sample, i.e., partial alignment of the magnetic moments in the direction parallel to the metallic glass ribbon surface. The D_{23} value after irradiation reduces to 2.2 and then further to 2.02, and the corresponding canting angles are 32.6° and 35°, respectively, which indicates reduction of in-plane anisotropy with the increase in irradiation. The increase in the average canting angle $\langle\theta'\rangle$ and other parameters fitted for Mössbauer spectra are given in table 1. The increase in average canting angle clearly indicates randomization of the magnetic moment for the sample irradiated with highest dose.

Similarly, for virgin $Fe_{40}Ni_{40}B_{20}$, $D_{23} = 3.11$ and $\langle\theta'\rangle = 20.7°$, indicating that the magnetic moments are aligned almost parallel to the ribbon surface. When the virgin sample is irradiated with the dose 5×10^{13} ions/cm^2, D_{23} reduces from 3.11 to 2.62 and canting angle increases to 27.1°. Further irradiation with higher dose 7.5×10^{13} ions/cm^2 reduces D_{23} to 2.52 and increases canting angle to 28.3°, showing reduction in in-plane magnetic anisotropy. The corresponding Mössbauer parameters are listed in table 2.

Table 1. Mössbauer parameters for $Fe_{40}Ni_{38}Mo_4B_{18}$, for virgin and the samples irradiated with 2 doses. $\langle H\rangle$ is the average hyperfine field, H_{peak} is the maximum field and ΔH is the width. D_{23} is the ratio of the 2,5 to the 3,4 spectral lines and D_{21} is the ratio of the 2,5 to the 1,6 spectral lines. $\langle\theta'\rangle$ is an average angle between the direction of magnetic moment and the ribbon plane and δ is the isomer shift.

$Fe_{40}Ni_{38}Mo_4B_{18}$	$\langle H\rangle$ (T)	H_{peak} (T)	ΔH (T)	D_{23} (±0.02)	D_{21} (±0.02)	$\langle\theta'\rangle$(±0.6) degrees	δ (mm/s)
Virgin	19.58	25	5.5	2.35	0.78	30.6	−0.102
5×10^{13} atoms/cm^2	18.21	23	5.8	2.2	0.73	32.6	−0.107
7.5×10^{13} atoms/cm^2	17.72	22	5.8	2.02	0.67	35	−0.103

Table 2. Mössbauer parameters for $Fe_{40}Ni_{40}B_{20}$, for virgin and the samples irradiated with 2 doses. $\langle H\rangle$ is the average hyperfine field, H_{peak} is the maximum field and ΔH is the width. D_{23} is the ratio of the 2,5 to the 3,4 spectral lines and D_{21} is the ratio of the 2,5 to the 1,6 spectral lines. $\langle\theta'\rangle$ is an average angle between the direction of magnetic moment and the ribbon plane and δ is the isomer shift.

$Fe_{40}Ni_{40}B_{20}$	$\langle H\rangle$ (T)	H_{peak} (T)	ΔH (T)	D_{23} (±0.02)	D_{21} (±0.02)	$\langle\theta'\rangle$ (±0.6) degrees	δ (mm/s)
Virgin	24.16	27	5.2	3.11	1.03	20.7	−0.0027
5×10^{13} atoms/cm^2	21.63	25	5.2	2.62	0.87	27.1	−0.1054
7.5×10^{13} atoms/cm^2	21.69	25	4.9	2.52	0.84	28.3	−0.119

Table 3. Irradiation dose and corresponding strain and residual stress, calculated for the samples $Fe_{40}Ni_{38}Mo_4B_{18}$ and $Fe_{40}Ni_{40}B_{20}$.

Sample	Dose (ions/cm^2)	Strain $\Delta x/x_0$	Residual stress $\sigma \pm 6$ (Gpa)
$Fe_{40}Ni_{38}Mo_4B_{18}$	5×10^{13}	0.543	33
	7.5×10^{13}	0.818	52
$Fe_{40}Ni_{40}B_{20}$	5×10^{13}	0.584	93
	7.5×10^{13}	0.885	141

Irradiation of metallic glasses with high-energy heavy ion beam results in the visible growth of sample dimensions, as seen by Klamunzer *et al* [6,17,18]. They have investigated an exhaustive number of metallic glasses of various compositions and in all cases, large dimensional changes have been found [6,17,18]. On irradiation with above incubation fluence B (which ranges from 0.6×10^{12} to 5.5×10^{12} ions/cm^2) metallic glasses exhibit dimensional change. The driving force of this phenomenon is the electronic energy loss S_e, which the ions deposit into electronic excitations and ionization [6,18]. In the model developed by Klamunzer [17], it is assumed that the passage of the ions through the sample generates locally high mechanical stresses, which release shear transformations. As a result, the atomic rearrangement occurs and dimensions perpendicular to the beam grow (i.e., the surface grows). Such atomic re-arrangements are possible only in metallic glasses because, unlike crystalline materials, they contain a large amount of free volume.

Here, we suggest that this effect can be responsible for the change in the orientation of magnetic moments in our samples. From the model, irradiation induced dimensional changes in the metallic glass can be given by the relation

$$\frac{\Delta x}{x_0} = A(\Phi t - B) \tag{3}$$

where x_0 is the original sample dimension and Δx is the change in the dimension after irradiation. $2A$ is the steady state growth rate, i.e., the change in the dimension per incoming ion, B is the incubation fluence and Φt is the fluence at which the sample is irradiated. The relation gives the strain produced due to the ion irradiation in metallic glasses. The data for the growth rate A, incubation fluence B and Young's modulus E for the metallic glasses under investigations are taken from [6,17,19]. Using the values of A and B in eq. (3), we determined the strain in our samples irradiated at different doses from which the residual stress could be easily calculated. Table 3 gives the calculated values of the strain and residual stress generated by the ion irradiation with the doses used in both the metallic glass samples.

When the calculated residual stress for the two different metallic glasses under investigation was linked to the change in average canting angle obtained by irradiation, a good degree of correlation was found. It is found that as residual stress increases the change in the average canting angle $\langle\theta'\rangle$ also increases. The data gives a straight line as a best fit as shown in figure 4. It can be safely inferred that the residual stresses produced in the glassy metals could be the main cause of the reduction in in-plane magnetic anisotropy. This phenomenon is in conformity with the magnetostriction effect in which mechanical stresses

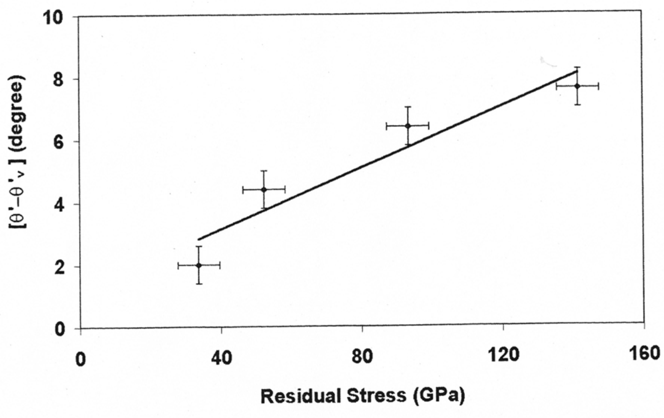

Figure 4. Change in average canting angle ($\theta' - \theta'_v$) vs. residual stress giving straight line as a best fit, for the irradiated samples $Fe_{40}Ni_{38}Mo_4B_{18}$ and $Fe_{40}Ni_{40}B_{20}$. θ'_v is average canting angle of the virgin sample.

induce magnetic anisotropy, except that the stresses introduced in the present work are not mechanically introduced but caused by the MeV ion irradiation.

4. Conclusion

It has been shown that the residual stresses introduced by electronic losses during high-energy ion irradiation in metallic glasses modify the magnetic anisotropy. A connection has been found between the effects of the dimensional change of metallic glasses and the modification of magnetic anisotropy, both induced by high-energy ion irradiation. It has been shown that the virgin samples of $Fe_{40}Ni_{40}B_{20}$ and $Fe_{40}Ni_{38}Mo_4B_{18}$ metallic glasses display in-plane magnetic anisotropy, i.e., the spins are oriented parallel to the ribbon plane. Irradiation is found to cause reduction in magnetic anisotropy and near complete randomization of magnetic moments has been observed at a dose of 7.5×10^{13} ions/cm^{-2}. It has been shown that there exists correlation between the residual stresses introduced by ion irradiation and the change in magnetic anisotropy in metallic glasses.

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