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Swift heavy ion induced surface and microstructural evolution in metallic glass thin films

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ABSTRACT

Swift heavy ion induced changes in microstructure and surface morphology of vapor deposited Fe–Ni based metallic glass thin films have been investigated by using atomic force microscopy, X-ray diffraction and transmission electron microscopy. Ion beam irradiation was carried out at room temperature with 103 MeV Au^{9+} beam with fluences ranging from 3×10^{11} to 3×10^{13} ions/cm^2. The atomic force microscopy images were subjected to power spectral density analysis and roughness analysis using an image analysis software. Clusters were found in the image of as-deposited samples, which indicates that the film growth is dominated by the island growth mode. As-deposited films were amorphous as evidenced from X-ray diffraction; however, high resolution transmission electron microscopy measurements revealed a short range atomic order in the samples with crystallites of size around 3 nm embedded in an amorphous matrix. X-ray diffraction pattern of the as-deposited films after irradiation does not show any appreciable changes, indicating that the passage of swift heavy ions stabilizes the short range atomic ordering, or even creates further amorphization. The crystallinity of the as-deposited Fe–Ni based films was improved by thermal annealing, and diffraction results indicated that ion beam irradiation on annealed samples results in grain fragmentation. On bombarding annealed films, the surface roughness of the films decreased initially, then, at higher fluences it increased. The observed change in surface morphology of the irradiated films is attributed to the interplay between ion induced sputtering, volume diffusion and surface diffusion.

1. Introduction

Metallic amorphous alloys are a class of materials that are widely investigated for plausible technological applications; owing to their enhanced structural, magnetic and electrical properties [1–4]. Most of these outstanding properties can be attributed to their random crystalline structure [3]. These materials can be synthesized by a variety of techniques like physical vapor deposition, solid-state reaction, ion beam irradiation, melt spinning, and mechanical alloying [3,4], of which, ion beam irradiation seems to be a promising technique since it can induce a variety of changes in the material ranging from bulk structural to local magnetic properties. During irradiation, swift heavy ions (SHI) passing through a material with velocities comparable to the Bohr velocity of electrons, lose energy mainly by two processes, namely, nuclear energy loss, and electronic energy loss. The former is dominant when the ion energy is in the keV range, and latter is dominant when the ion energy is in the MeV range.

Most of the effects associated with SHI irradiation occur in the electronic energy loss regime. This inelastic scattering assisted energy loss of fast heavy ions creates latent tracks, phase transitions, amorphization, damage creation, annealing effects, dimensional changes, and nanostructures. During irradiation, the surface morphology also evolves resulting from the competition between dynamic roughening; which increases the surface roughness, and smoothening which decreases it. Recently, A. Kanjilal and D. Kanjilal [5] reported surface roughening kinetics of 100 MeV Au beam irradiated $Si_{1-x}Ge_x$ alloy films for $x = 0.5$ and 0.7. The irradiation induced surface roughening behavior is demonstrated by studying the variation of surface roughness as a function of fluence. The composition

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dependent variation of surface morphology with increasing fluence is ascribed to the strain distribution along the sample surface. Thomas et al. [6] reported SHI irradiation induced coercivity changes in Fe–Ni based thin films and the observed changes were correlated with topographical evolution of the films with fluence. Dash et al. [7] carried out quantitative roughness and microstructure analysis of as-deposited and SHI (107 MeV Ag and 58 MeV Ni) irradiated 10 and 20 nm thick Au films using AFM and its power spectral density analysis. They reported an increase in the root mean square (rms) roughness at low fluences and a decrease at higher fluences. The PSD analysis also showed similar variation of low frequency roughness with ion fluence. In the high frequency regime, surface morphology of irradiated samples was found to be governed by a combined effect of evaporation–re-condensation and diffusion dominated processes. Gupta and Avasthi [8] reported sputtering in Au thin films, resulting from energy deposition in the films, owing to inelastic collision of SHI with electron clouds of target atoms. Gupta et al. [9] reported SHI induced surface smoothening, roughening and sputtering of thermally immiscible Fe/Bi bilayer system. The observed behavior of surface smoothing and roughening under SHI irradiation was explained on the basis of the thermal spike model. In general, magnetic properties of thin films are sensitively correlated to the surface roughness and hence it will be worthwhile to probe possible mechanisms by which roughness of thin films can be tailored. Further, rough films can act as templates for the growth of nanostructures using oblique angle deposition [10]. Pre-patterned substrates offer a suitable platform for growth of nanostructures by suitable deposition techniques.

There are also reports that electronic energy transfer to the amorphous alloys can lead to anisotropic growth, which means, shrinkage of alloy ribbons in the direction of the beam and expansion in the direction transverse to it [11]. In thin films, this type of effects can create large stress in the film-substrate interfacial boundaries. The dense random arrangement of atoms in metallic glasses favors amorphous structure due to the increased liquid–solid interfacial energy. Any structural or morphological changes occurring in these materials are of great importance in determining the magnetic properties. Hence, amorphous alloys can serve as ideal templates to investigate these varied effects.

Scanning probe microscopy (SPM) is a versatile technique to probe surface morphology of thin films in the nano range. In case of magnetic thin films, surface morphology from AFM and magnetic morphology from magnetic force microscopy (if there is considerable out-of-plane stray field) can be extracted employing specific tips in SPM. The images can be further subjected to Fourier analysis using image analysis software to obtain PSD, roughness and auto correlation functions.

Most of the available literatures pertaining to the study of material modification by ion beams were based on bulk materials. Work on metallic glass thin films based on iron and nickel is seldom reported [12–16]. The authors group recently reported 108 MeV Ag^{8+} ions induced surface modification of Fe–Ni based metallic glass thin films in the as-deposited state [6]. In the present investigation, we focus on the influence of swift heavy ions on the microstructure and surface morphology of 673 K annealed metallic glass thin films, so as to study the impact of SHI on the crystalline nature of these films. The results are correlated with a view to gain insight into the structural and morphology evolution with SHI irradiation.

2. Experimental

2.1. Thin film preparation

Metglas thin films with nominal thickness of 100 nm were vacuum evaporated using tungsten filaments on chemically and ultrasonically cleaned glass substrates from a composite target having composition $Fe_{40}Ni_{38}B_{18}Mo_4$. The chamber pressure before deposition was 1×10^{-6} mbar, which increased to 3×10^{-5} mbar during deposition. We recently studied the microstructure and magnetic evolution of Fe–Ni based thin films with thermal annealing [12,14]. From the TEM images it was found that, the microstructure of as-deposited films exhibits a contrast typical of an amorphous material [14]. The bright field TEM images of the films annealed at 473, 573 and 673 K, revealed that the microstructure consisted of nanocrystallites embedded in an amorphous phase. Grain growth was also observed with an increase in the annealing temperature [12,14]. Based on those results, the as-deposited Fe–Ni based metallic glass films were annealed at 673 K for 1 h. Annealing was performed at 4×10^{-5} mbar for minimizing surface oxidation.

2.2. Swift heavy ion irradiation

As deposited as well as annealed films were irradiated with 103 MeV Au^{9+} ions at the 15 UD Pelletron accelerator at IUAC, New Delhi. The irradiations were performed at 0° angle of incidence with respect to the surface normal. Ion beam was raster scanned on the sample surface by a magnetic scanner for maintaining a uniform ion flux throughout the film. The fluences were varied from 3×10^{11} to 3×10^{13} ions/cm^2. The irradiated sample area was 1 cm^2. The Au ion is chosen due to its higher mass and the energy regime is selected after simulation using SRIM code [17]. For the chosen ion energy of 103 MeV, the lateral straggling is 5.87 μm, longitudinal straggling is 4.62 μm and the penetration depth is 6.98 μm. This value of penetration depth is two orders of magnitude greater than the thickness of the film. The energy of the Au^{9+} beams (103 MeV) was selected with a view to avoid the ion implantations in the film with maximum electronic loss of 39 keV/nm and minimum nuclear energy losses within the accelerators maximum energy limit (see Fig. 1).

The samples were mounted on a massive copper block using carbon tape. The increase in sample temperature during ion irradiation can be estimated using the Fourier heat conduction equation, $j = -\lambda \frac{dT}{dz}$. The total heat carried into the system can be taken as the total energy carried by SHI, = ϕE_{shi}, where ϕ is the ion flux and E_{shi} is the ion energy. In this study, a low ion flux of 6×10^9 cm^2/s was maintained with energy of 103 MeV per ions. The conductivity of float glass is nearly equal to 1 W m^{-1} K^{-1}. However, even if we

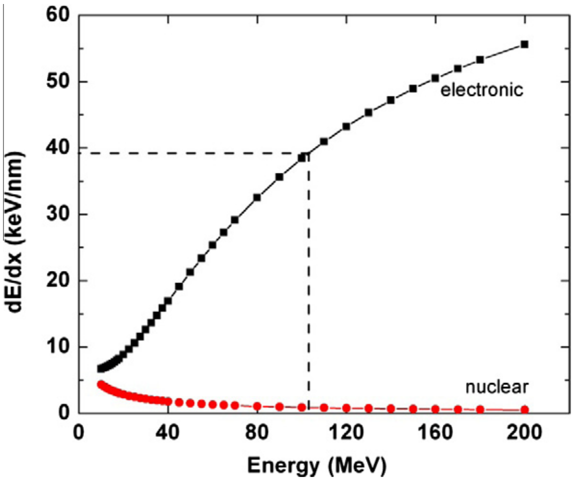

Fig. 1. SRIM simulation showing electronic and nuclear energy loss versus ion energy. Dashed line shows the electronic energy loss corresponding to 103 MeV energy.