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Study of low energy Ar^+ ion irradiated $^{57}Fe/Pt$ multilayers

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Abstract

This paper reports a study on the effects of Ar^+ ion irradiation in $^{57}Fe/Pt$ multilayers having the structure Si (substrate)/[^{57}Fe(19 Å)/Pt(25 Å)]$_{\times 10}$. The as-deposited multilayers were irradiated with 90 keV Ar^+ ions at a fluence of 2×10^{16} ions cm^{-2} at different temperatures. X-ray reflectivity, x-ray diffraction and conversion electron Mossbauer spectroscopy were used for the characterization of the irradiated samples. Using x-ray reflectivity it was observed that irradiation results in mixing across the interfaces and deterioration of the multilayer structure. The mixing results in the formation of fcc FePt phase and no $L1_0$ ordering is observed in the samples. The results are discussed in terms of recoil displacements induced, energy transfers from ions supporting the observations of Bernas *et al*, using kinetic Monto Carlo simulations.

1. Introduction

Ion beam mixing is a powerful tool for modifying the surfaces and interfaces at a nanometre scale. The effects of an ion beam in solids are different depending on the energy. Ion beams having energies ∼MeV/nucleon lose their energy in the target via inelastic collisions involving excitation and ionization of the target atoms, and ion beams having energies ∼keV/nucleon lose energy in the target via elastic collisions [1]. Depending on the type of ion, its energy and fluence, ion bombardment can promote local atomic rearrangement and can modify the microstructure of alloys. These modifications are strongly dependent on the details of the chemical interaction between the constituents, their miscibility and heats of mixing. In most of the bilayer systems composed of miscible elements (having negative heat of mixing), the

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measured mixing rates were up to ten times larger than those predicted ballistically and were found to depend on the thermodynamic properties like heat of mixing and cohesive energy [2]. The FePt system is considered to be technologically important, because of its huge magnetic anisotropy values ($K_u \sim 10^7$ erg cm^{-3}). The transformation of the disordered face-centred cubic (fcc) phase to the ordered face-centred tetragonal (fct) $L1_0$ phase has been attributed to be the reason for the high magnetic anisotropy [3]. The phases that will be formed due to intermixing in the Fe/Pt system include the equi-atomic disordered fcc FePt, ordered fct FePt, iron-rich Fe_3Pt and platinum-rich $FePt_3$ phases. Using low energy light ions, an improvement in the ordering of FePt has been reported in the literature [4–6]. The aim of the present paper is to understand the role of recoil displacements, damage creation with the ion irradiation in the formation of the ordered fct phase of FePt, by studying the effects of Ar^+ ion irradiation at different temperatures in a system having the structure Si (substrate)/$[^{57}$Fe(1.9 nm)/Pt(2.5 nm)$]_{\times 10}$.

2. Experimental details

$[^{57}$Fe(1.9 nm)/Pt(2.5 nm)$]_{\times 10}$ multilayers were deposited on silicon substrate using ion beam sputtering. It may be noted that the thickness is selected such that to have an equal number of atoms. A beam of argon ions (purity of 99.9995%) was used to sputter ^{57}Fe-enriched Fe and Pt targets using a Kaufman-type hot-cathode ion source. A base vacuum of 1×10^{-7} Torr was achieved before deposition. The targets were mounted on a rotary motion feed through to switch over from Fe to Pt in order to deposit alternate layers. The deposition rates for Fe and Pt were 0.05 and 0.07 nm s^{-1} respectively. The as-deposited samples were irradiated with 90 keV Ar^+ ions at a fluence of 2×10^{16} ions cm^{-2} at different temperatures, namely, 125, 189 and 325 °C (henceforth designated as Ar125, Ar189 and Ar325 respectively; the as-deposited sample is designated as Ar0) using an 150 kV ion accelerator at IGCAR, Kalpakkam, India. For the FePt alloy, the critical temperature (T_c) for the radiation enhanced diffusion (RED) process calculated using the empirical relation $T_c = 95.2^* \Delta H_{coh}$ (eV/atom) [7, 8] is as 208 °C. The above irradiation temperatures were chosen so that the irradiation was carried out below, close to and above T_c. The ion beam current density was 0.5 μA cm^{-2} and the vacuum level at target chamber was 1×10^{-6} Torr. The temperature during the irradiation was measured using a chromel–alumel thermocouple. The energy was selected such that the range of the ion covers the total thickness of the film and causes significant atomic displacements in the system [9]. The x-ray reflectivity (XRR) and grazing incidence x-ray diffraction (GIXRD) measurements (the angle of incidence 0.5°) were carried out for both as-deposited and irradiated samples using a Siemens D5000 diffractometer with Cu Kα radiation. The magnetic measurements were carried out using the conversion electron Mossbauer effect (CEMS).

3. Results and discussion

Figure 1 represents the x-ray reflectivity patterns of the as-deposited and irradiated multilayers. The Bragg peaks due to the multilayer periodicity up to third order demonstrate the clear stratification in the as-deposited sample. The XRR patterns were fitted with the Parratt formalism [10]. The solid line represents the best fit to the data. The obtained bilayer periodicity from XRR is 4.48 nm. As is seen from figure 1, the third-order Bragg peak disappears with irradiation. And it is also observed from the patterns that with irradiation the height of the first Bragg peak decreases. The reduction in the height of the first Bragg peak indicates the interdiffusion across the layers of the multilayer. It is also observed that with irradiation the interface roughness increases. The increase in the interface roughness is

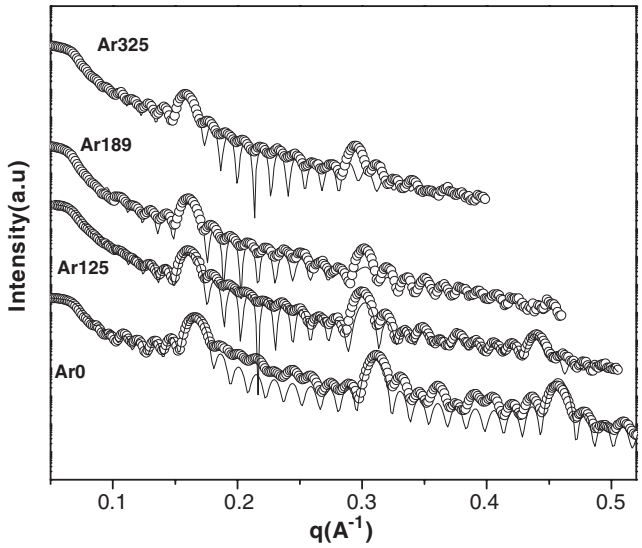

Figure 1. XRR patterns of the as-deposited and irradiated multilayers. The x-axis is the momentum transfer $[q = (4\pi \sin\theta)/\lambda]$ and the y-axis is the intensity plotted on a log scale.

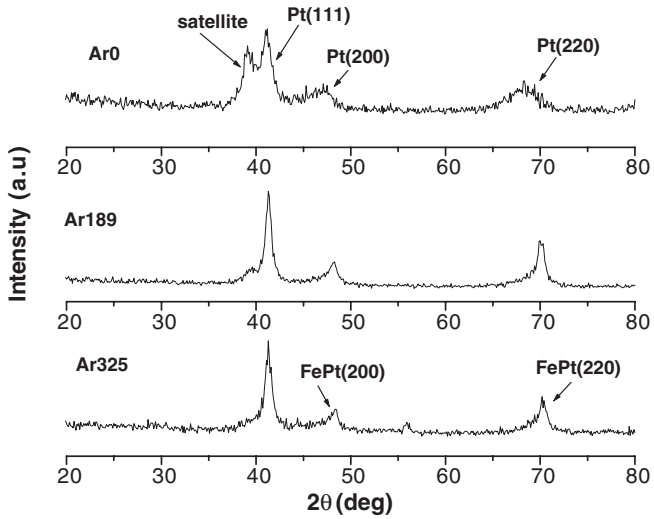

Figure 2. GIXRD patterns of the as-deposited and irradiated multilayers. The intensity is plotted on a linear scale.

considered to be equivalent to that due to the formation of alloy at the interfaces. Figure 2 shows the GIXRD patterns of the as-deposited and irradiated multilayers. The GIXRD pattern of the as-deposited film shows two sharp peaks at 39.17° and 41.13°. The crystalline size calculated from the width of the peak at 41.13° using the Debye–Scherrer formula [11] is 8.82 nm. This indicates that there is strong coherence between the Fe and Pt layers of the multilayer, and at the same time the Mossbauer results as discussed below show a smaller BHF value indicating that considerable amount of Pt has interdiffused into the Fe layers. The high angle x-ray patterns were fitted with the superlattice refinement (SUPREX) program [12]

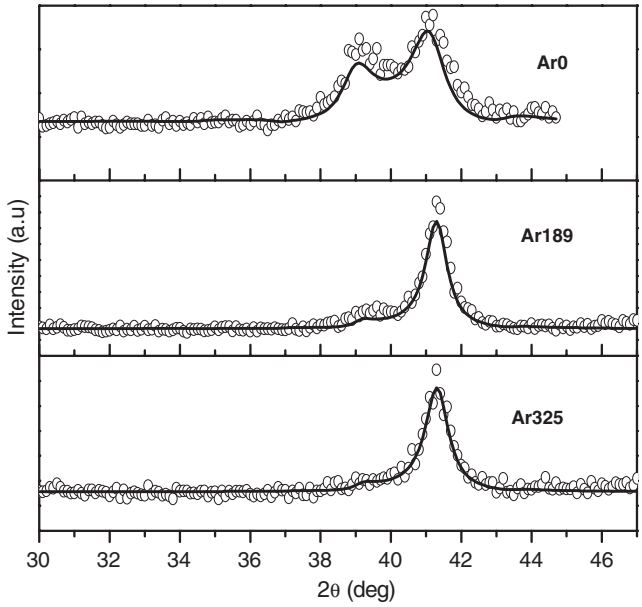

Figure 3. High angle x-ray patters of the as-deposited and irradiated multilayers. The solid line represents the best fit to the data using SUPREX. The intensity is plotted on a linear scale.

with the interdiffusion model [13]. Figure 3 shows the high angle x-ray profiles along with the fitted data. The obtained parameters from the SUPREX fitting are N_{Fe} and N_{Pt} as 6.1104 and 13.4108; d_{Fe} and d_{Pt} as 0.207 10 and 0.224 92 nm respectively (N and d are the number of atomic layers and crystalline lattice spacing respectively), giving the bilayer periodicity as 4.28 nm, which matches closely with the value obtained from XRR. Keeping all the parameters fixed, the interface width (the interface widths of Fe–Pt and Pt–Fe are assumed to be equal) was varied to fit the data of the irradiated spectra. The interface width was observed to increase from 3.27 to about 16 monolayers for the Ar325 sample. It is observed that as the irradiation temperature increases, the satellite peak disappears. This is due to the diffusion of atoms across the interfaces. The observed shift of the Pt(111) peak position in the as-deposited sample is similar to the case of sputter deposited Co/Au multilayers [14]. In the as-deposited sample two broad peaks are observed at 46.63° and 68.19°, which are due to the Pt(200) and Pt(220). With the high temperature irradiation, these broad peaks are resolved into sharp peaks at 48.0° and 70.0°. These are identified as disordered fcc FePt(200) and FePt(220) respectively. The peak at 55.9° in the Ar325 sample might be due to Si(311). Therefore, the low and high angle x-ray diffraction results indicate intermixing and formation of FePt alloy in the multilayers with high temperature irradiation. Figure 4 shows CEMS spectra of the as-deposited and irradiated films. The Mossbauer spectrum was fitted using the NORMOS-DIST program (http://www.wissel-instruments.de/produkte/software.html). It is observed that as the temperature is increased, a central doublet is observed in the CEMS spectra. The normalized hyperfine field distribution of all the samples is shown figure 5. It may be noted that as the temperature is increased, the intensity of the peak at lower field values increases systematically. This might be due to the non-magnetic amorphous iron silicide formation at the interface of the silicon substrate and first Fe layer [15]. An average hyperfine field of 24.7 T is observed in the as-deposited film. This is because the neighboring platinum atoms around the iron atoms

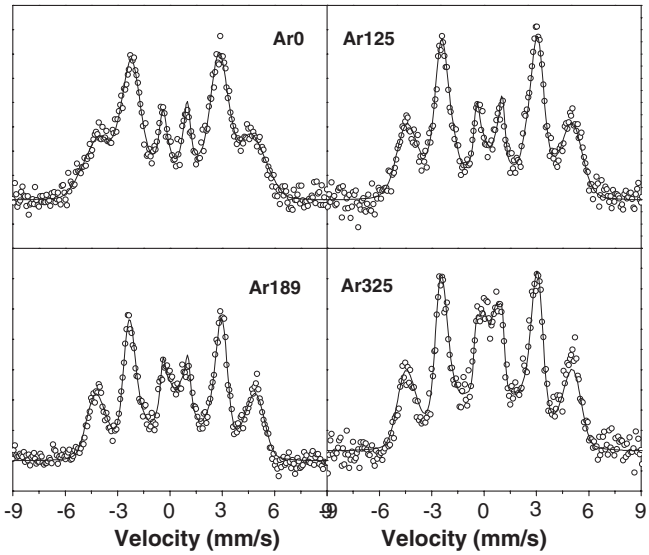

Figure 4. CEMS spectra of as-deposited and high temperature irradiated samples.

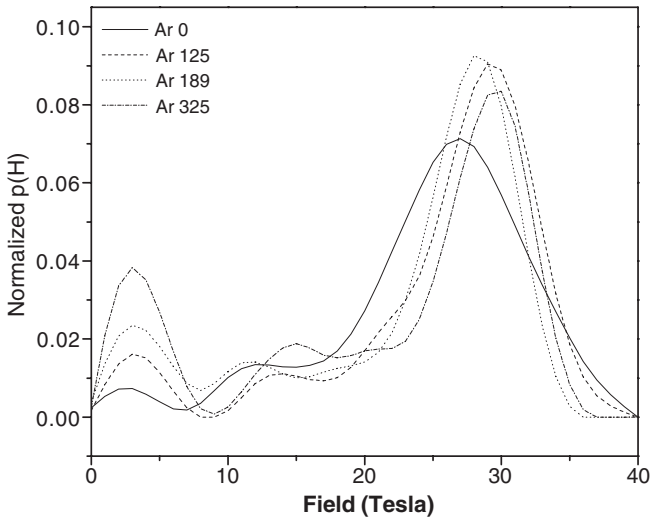

Figure 5. Normalized hyperfine field distribution obtained from Mossbauer spectra analysis.

reduce the hyperfine field of Fe [16], as some amount of Pt is expected to diffuse into the Fe layer in the as-deposited film. The spectrum of the Ar325 sample still shows very broad lines, which must be due to the disordered cubic FePt phase. No quadrupole splitting is observed in the Ar325 sample indicating the formation of fcc phase of FePt and an A23 (i.e., area ratio of the second and third Mossbauer lines) value of 3.75 indicates that the magnetization is in the plane of the film. Therefore, from the above studies one can conclude that with high temperature 90 keV Ar^+ irradiation mixing will result in Fe/Pt multilayers leading to the formation of the fcc FePt phase.

The irradiation effects in the FePt system using 130 keV and 2 MeV He^+ ions have been studied and reported to have enhanced ordering [5, 6]. In both the above studies the

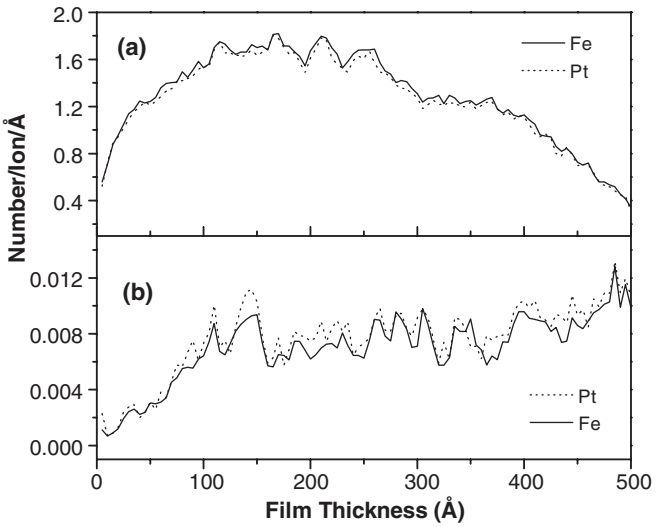

Figure 6. Damage events (vacancies) calculated from SRIM code for FePt film (a) with 90 keV Ar^+ and (b) with 130 keV He^+ ions.

temperature during irradiation was above T_c and the improvement in the ordering can be understood as one expects ordering at a lower temperature in accordance with the equation $T' = T(1 + \Delta)$, where T is the temperature at which irradiation is carried out, and Δ is the dilatation parameter which is a function of irradiation fluence, temperature and material properties [17]. However, the absence of ordering in our present study even though the irradiation was carried out above T_c can be due to the production of large vacancies (a quick estimation of the damage events for the two ion species using Monte Carlo simulation SRIM-2003 is shown in figure 6) and large recoil displacements with the Ar^+ ion as compared to the He^+ ion. Therefore, the present observations support the findings of Bernas *et al*, using kinetic Monte Carlo simulations wherein they have concluded that when irradiation induced rearrangements are on a near-neighbour scale then successive pairwise interactions lead to efficient ordering [4].

In addition to the above arguments, because of the multilayer structure reduced dimensionality might be playing a role in not promoting the $L1_0$ phase. Matsumura *et al* [18] have not observed any promotion of $L1_0$ ordering in FePt nanoparticles embedded in an Al_2O_3 matrix with 100 keV He^+ ion irradiation at high temperature, and concluded that the ordering tendency of nanoparticles is reduced owing to the size effect.

In summary, we have studied the structural and magnetic properties of $[^{57}Fe(19\ Å)/Pt(25\ Å)]_{\times 10}$ multilayers irradiated above and below the critical temperature of the radiation enhanced diffusion process with 90 keV Ar^+ ions. With the irradiation an intermixing in multilayers was observed, leading to the formation of the fcc phase of FePt. No superstructure fct phase of FePt was observed with the irradiation. The results of the present experiment can be understood in terms of recoil displacements induced and energy transfers from ions. The present results along with the light ion irradiation supports the observations of Bernas *et al*, using kinetic Monto Carlo simulations, in which it was shown that when irradiation induced rearrangements are on a near-neighbour scale, then successive pairwise interactions lead to efficient ordering.

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