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Si^+ ion irradiation in a Co/Pt multilayer system

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Abstract
This paper deals with a study of the effect of Si^+ ion irradiation on a Co/Pt multilayer system irradiated at different temperatures. The as-deposited and irradiated samples have been characterized using x-ray reflectivity (XRR), x-ray diffraction (XRD) and the magneto-optical Kerr effect (MOKE). X-ray reflectivity shows clear intermixing at the interfaces. The x-ray diffraction pattern shows that Si^+ ion irradiation at higher temperatures results in the formation of the $CoPt_3$ fcc phase with a small fraction of $L1_0$ phase. The mixing process is discussed in terms of recoil displacements induced by energy transfers from ions.

1. Introduction

The CoPt system has been extensively studied due to its fascinating magnetic properties. CoPt alloy thin films possess high anisotropy and coercivity [1]. CoPt alloy thin films are potential candidates for magnetic devices like high density recording media and magnetic bias films of magneto resistive elements. Co/Pt multilayers exhibit structure-sensitive magnetic anisotropy and giant magnetoresistance (GMR). Co/Pt multilayer thin films have a large of coercive field to saturation magnetization ratio. Co/Pt multilayers are found to exhibit large coercivities in both sputtered and evaporated films [2]. In the as-deposited state, the CoPt system is in a chemically disordered phase where Co and Pt on the fcc lattice are randomly distributed, and hence exhibits weak magnetic properties. It has been reported that with thermal annealing or ion irradiation, the system transforms to the ordered fct phase with excellent magnetic properties [3]. However, the annealing temperature required to induce ordering in the CoPt system is very high, which also results in substantial grain growth. Such grain growth is undesirable, as high density recording requires a bit dimension of only a few nanometers in diameter. Recently ion irradiation has been found to be an effective tool in tailoring the structural and magnetic properties of any system. The advantages of ion irradiation are to introduce an appropriate number of atomic species and desired energy into the films, which may create defects or lattice distortion and may thus promote phase transformation at lower temperatures. The ion-beam mixing process is known to be able to break atomic bonding to mobilize atoms, which results in the formation of a mixed-alloy phase. The energy transfer of the incident ions can heat the sample to high temperatures (several 10^4 °C) in a localized region with cooling down to room temperature (RT) on a timescale of 10^{-8} s. Hence, this technique enables us to make various metastable compounds, regardless of the kind of elements and the atomic stoichiometry [4]. While conventional annealing allows us to obtain an intermetallic compound only at much higher temperature than RT, ion-beam mixing is capable of preparing alloy films at RT. In particular we can obtain a metastable solid phase, which does not exist in a thermodynamic equilibrium state.

It has been reported that ion irradiation has been employed to tune the anisotropy direction of CoPt [5], and to manipulate the magnitude and the direction of the exchange field [6]. The magnetic properties of the CoPt ultra-thin structure with focused Ga ion irradiation in the energy range 20–100 keV have been reported [7]. It has been observed that at lower fluences (2×10^{14} ions cm^{-2}) the coercive field decreases slowly with irradiation fluence, and at higher fluences collisional intermixing induces significant alloying of the Co layer and the film turns paramagnetic at room temperature. However, Chang *et al* [8] showed that magnetic anisotropy could be induced in a ferromagnetic Co/Pt multilayer film by employing Ar ion irradiation in an externally applied magnetic field. Balaji *et al* [9] observed $L1_0$ phase formation

in self-ion implanted CoPt bilayers followed by high energy Si ion irradiation at elevated temperatures. It would be interesting to study the ion-induced modifications in a CoPt multilayer system with Si ion irradiation at different substrate temperatures. The aim of the present paper is to understand the phase formation mechanism and the role of radiation enhanced diffusion with Si ion irradiation by making use of recoil displacements.

2. Experimental details

[Co (12Å)/Pt (17Å)] $\times 15$ multilayers were deposited on float glass using ion beam sputtering. The rate of deposition for Co and Pt was 38 Å and 42 Å min^{-1}, respectively. It may be noted that the layer thicknesses are selected so as to have approximately equal number of Co and Pt atoms. A beam of argon ions (purity of 99.9995%) was used to sputter Co 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 feedthrough to switch over from Co to Pt in order to deposit alternate layers. Different pieces of as-deposited samples were irradiated with 4 MeV Si^{+} ions to a fluence of 2×10^{16} ions cm^{-2} at different temperatures, namely 200, 250, 300 and 350 °C, using the 1.7 MV Tandetron accelerator at IGCAR, Kalpakkam, India. The energy has been selected such that range of the ions covers the total thickness of the film and causes significant atomic displacements in the system (from the calculation of the stopping range of ions in matter, the range was found to be 1.09 μm) [10]). The ion beam current was set to 0.5 μA cm^{-2} and the total irradiation time was taken to be 5 h, so as to have a fluence of 2×10^{16} ions cm^{-2}. A vacuum of the order 1×10^{-6} Torr was maintained during irradiation. The temperature of the sample was measured using a chromel–alumel thermocouple and it did not rise. The irradiated samples were analyzed by x-ray reflectivity (XRR), wide angle x-ray diffraction (XRD) and the magneto-optical Kerr effect (MOKE).

3. Results and discussion

Figure 1 shows the XRR pattern of the as-deposited and irradiated films at different temperatures. The reflectivity pattern for the pristine sample shows prominent Bragg peaks up to second order, which appear due to the well ordered periodic nature of the Co/Pt multilayer. Simulation of the XRR of pristine and irradiated samples was performed on a multilayer model-dependent approach following Parratt's formalism [11]. The solid line represents the best fit to the data. The bilayer periodicity of the pristine film was found to be 25 Å instead of the designated 29 Å. This may be because of some variation in deposition conditions. Upon irradiation the heights of both first and second order Bragg peaks reduce gradually. This indicates increasing intermixing as a function of irradiation temperature. As expected, the height of the second Bragg peak decays at a faster rate with intermixing [12–14]. However, even after irradiation at the highest temperature of 350 °C, the first order Bragg peak does not disappear completely, indicating that the system is not yet completely intermixed. One may note that there is variation in the position of the first Bragg peak which is due to the fact that different pieces of pristine sample (having some thickness variation) were irradiated at different temperatures.

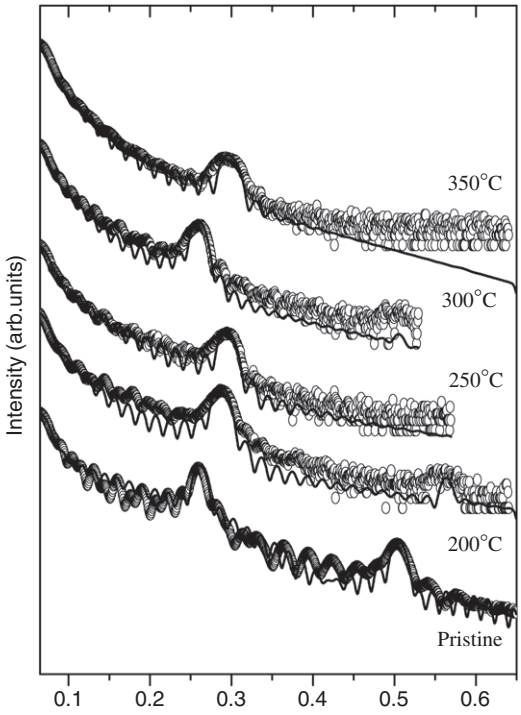

Figure 1. XRR patterns of the as-deposited and irradiated multilayers. The x-axis is the momentum transfer [$q = (4\pi \sin\theta)/\lambda$] and the y-axis is the intensity plotted on a log scale.

Figure 2 shows the XRD of the Co/Pt samples irradiated at different temperatures. The pristine sample shows an intense peak at 41.14°. We ascribe this peak to the Pt(111) reflection. It has been found that in Fe/Pt, Co/Pt, Cu/Au multilayer systems the position of the Pt (Au) peak gets shifted to a higher angle because of stress induced by the neighboring Fe (Co, Cu) layers [15–17]. The stress is tensile in the Pt layer and compressive in the Co layer. Using the Scherrer formula, the coherence length was found to be 68.5 Å, which is greater than the bilayer periodicity. This suggests that there is some degree of coherency between Pt and Co layers. There is a small peak at 37.28° around the Pt(111) peak, which is known as a satellite peak arising due to the regular periodic arrangement of bilayers. Similar results were observed in our earlier studies [18]. The high angle x-ray pattern of the pristine sample has been fitted with the superlattice refinement (SUPREX) program [19] with the interdiffusion model [20]. Figure 3 shows the high angle x-ray profile along with the fitted data. The obtained parameters from the SUPREX fitting are N_{Co} and N_{Pt} as 4.898 and 6.578, respectively, d_{Co} and d_{Pt} as 0.215 and 0.222 nm, respectively, where N_{Co} and N_{Pt} denote the number of atomic planes of Co and Pt, while d_{Co} and d_{Pt} denote their interplanar distances. The bilayer periodicity (Λ) has been calculated using the relation

$$\Lambda = N_{\text{Co}} d_{\text{Co}} + N_{\text{Pt}} d_{\text{Pt}}, \tag{1}$$

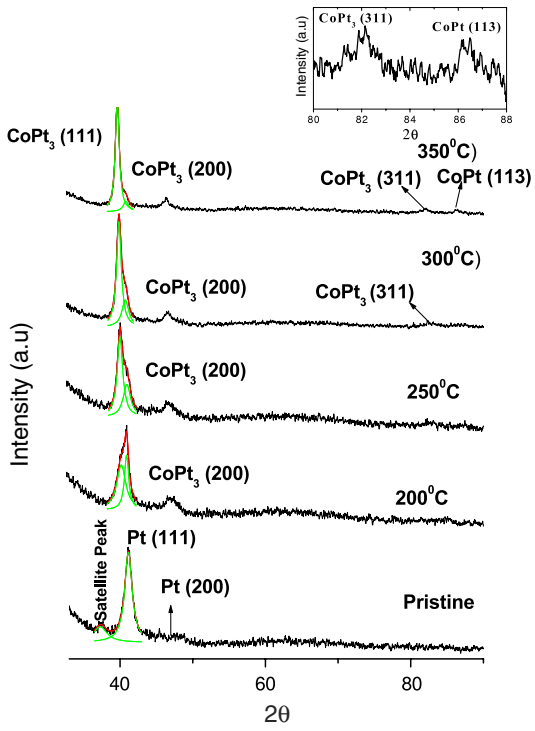

Figure 2. XRD patterns of the as-deposited and irradiated multilayers. The intensity is plotted on a linear scale.
(This figure is in colour only in the electronic version)

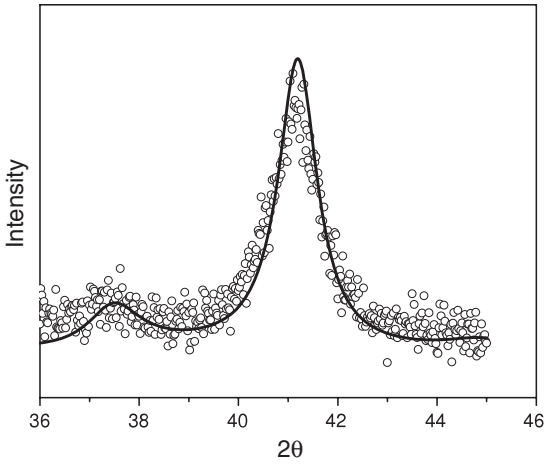

Figure 3. High angle x-ray patterns of the as-deposited Co/Pt multilayer. The solid line represents the best fit to the data using SUPREX. The intensity is plotted on a linear scale.

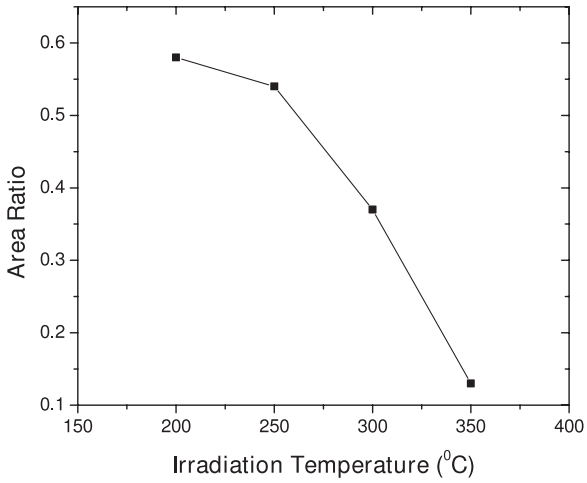

Figure 4. Variation of the area ratio of the Pt(111) peak to the fcc $CoPt_3$(111) peak with high temperature irradiation.

and was found to be 2.5 nm, which matches well with the XRR results. It is observed that the satellite peak disappears with irradiation at a temperature of 200 °C and above. This is due to the diffusion of atoms across the interfaces. After irradiation, the Pt(111) peak gets transformed to a $CoPt_3$(111) peak with a slight shift to a lower angle. Chang *et al* [21] reported a shift of 0.4° in the (111) peak position of the CoPt phase to lower angles in Ar^+ ion irradiated Co/Pt multilayers, and have attributed it to expanded lattice spacing. In the present experiments we have found a shift of 0.5° in the (111) peak position of the $CoPt_3$ fcc phase to a lower angle with irradiation, indicating that the planar spacing of the metastable state is slightly increased. Further, it has been observed that with irradiation at high temperatures, the (111) $CoPt_3$ peak was a broad peak formed by an overlap of two peaks, one corresponding to the $CoPt_3$(111) fcc peak and the other corresponding to the Pt(111) phase. Accordingly, the XRD patterns in the angular range $36° < 2\theta < 44°$ have been fitted using non-linear curve fitting with two overlapping Lorentzian peaks. With irradiation at 200 °C, the coherence length was still found to be greater than the bilayer periodicity, indicating that there is still coherency between the Co and Pt grains. It has been observed from the fitting that the area of the $CoPt_3$ fcc phase is increasing at the expense of the Pt(111) peak. Figure 4 shows a plot of the area ratio of the Pt(111) peak to that of the $CoPt_3$(111) fcc peak. Other peaks corresponding to fcc $CoPt_3$(200), (311) have been observed in the irradiated samples. One may note that even after irradiation at a temperature of 350 °C some amount of Pt(111) peak is left, suggesting that intermixing of Co and Pt layers is not complete. This is in agreement with our XRR results, where a small Bragg peak in reflectivity persists even after irradiation at 350 °C. However, in the sample irradiated at 350 °C we could observe the evolution of CoPt(113) peaks which correspond to the CoPt fct phase (the inset in figure 2 shows the evolution of the fct CoPt(113) peak). This suggests that along with the presence of some fcc phase there is a fraction of ordered phase present in the system. It may be noted that with irradiation at elevated temperatures there might be the possibility of diffusion of Na atoms into the Co/Pt multilayer due to heating. However, this diffusion would be limited to one or two layers of the film, and would not have any significant effect on the phase formation mechanism and ordering process. It is observed that the crystallite size corresponding to the $CoPt_3$ phase is increasing with irradiation, which indicates that with irradiation there is some mixing observed in the system in such a manner that there are some grains of $CoPt_3$ and some Pt and Co grains with a strong coherency. It has been observed that Si ion irradiation in a Co/Pt multilayer

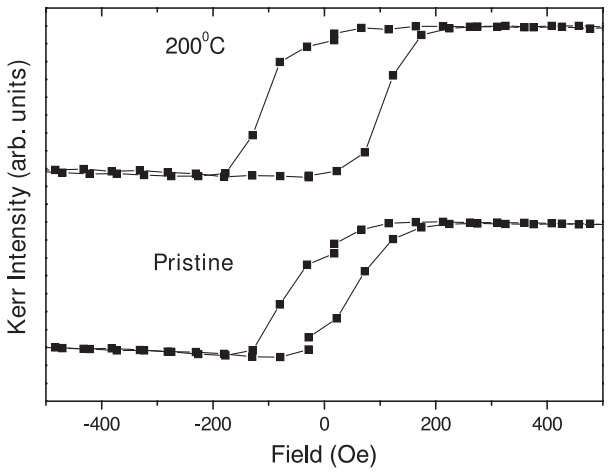

Figure 5. Hysteresis loops of as-deposited and irradiated Co/Pt multilayers.

system leads to the formation of a non-equilibrium phase. This can be explained in the following manner: when energetic ions of several tens to hundreds of keV penetrate a thin film at elevated temperatures, a thermally activated long-range diffusion process sets in when defects generated by the incoming ions become mobile; this is known as radiation enhanced diffusion (RED). The critical temperature for RED, T_c, is defined as the temperature at which the temperature-dependent mixing equals the temperature-independent cascade mixing. This process may cause displacements of the target ions, producing a new mixed phase through mixing of the cobalt and non-platinum sublayers. The studies demonstrate that these alloys have a slightly expanded unit cell and reduced coordination that greatly reduces the electronic interaction between atoms in comparison with the stable alloy [22]. The metastable conditions are the result of the rapid ($\sim 10^{-11}$ s) quenching corresponding to an effective cooling rate of 10^{14} K s^{-1}.

The hysteresis loops were recorded at room temperature by measuring the Kerr rotation (MOKE) with the field applied in the plane of the sample. The MOKE pattern is shown in figure 5. The pristine sample exhibits a loop with very little coercivity. The pattern shows that there is a considerable increase in coercivity in the sample irradiated at a temperature of 200 °C. The coercivity for the pure unirradiated Co/Pt multilayers is about 50 Oe, which increases to about 100 Oe for the irradiated sample. This change is associated with the mixing of Co and Pt layers of fcc symmetry. In the sample irradiated at 250 °C it is found that anisotropy has developed in the system and the loop was not able to saturate with the present existing field of 1.8 kOe.

The critical temperature (T_c) of CoPt alloy for the RED process is calculated by using the empirical relation $T_c = 95.2 H_{coh}$ (eV/atom) [23, 24], and found to be 214 °C, where H_{coh} is the cohesive energy. The Si^+ ion irradiation was carried out at temperatures starting from 200 to 350 °C, which is well above the T_c for RED to occur. It has been observed that with high temperature irradiation, mixing is taking place across the interfaces as confirmed by XRR and XRD. To initiate ordering in the target material, the irradiating beam should have: (a) small energy transfers, (b) minimized recoil displacements, and (c) a low collision cross section in order to avoid defect interactions [25]. The energy absorbed by the recoils in CoPt film from 4 MeV Si^+ ions is 50.36 eV/incoming ion/nm, and the number of displaced atoms is 60.95×10^{-2}/incoming ion/nm, as observed from stopping range ions in matter calculation (SRIM) [10]. These recoil atoms lead to mixing across the interfaces. The low energy transfers and small number of displaced atoms produced by 4 MeV Si^+ ion irradiation enables the mixing process in the system, giving rise to the formation of the fcc $CoPt_3$ phase and a small fraction of $L1_0$ CoPt phase.

4. Conclusions

The structural and magnetic properties of the Co/Pt system with Si^+ ion irradiation have been studied. It has been observed that with Si ion irradiation mixing has been observed in the Co/Pt multilayer system. The XRD pattern reveals that ion irradiation leads to the formation of a non-equilibrium fcc $CoPt_3$ phase along with a fraction of $L1_0$ phase.

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