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# Realization of magnetic anisotropy and $L1_0$ CoPt ordered phase by Pt<sup>+</sup> ion irradiation on a Co/Pt bilayer film

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# Abstract

Ion-beam synthesis of a CoPt layer was achieved by Pt<sup>+</sup> ion irradiation on Pt/Co bilayer films, and further growth and ordering of the CoPt layer was seen upon 4 MeV Si<sup>+</sup> ion irradiation at 300 °C. Rutherford backscattering (RBS) results show that the ion-beam mixing increases as a function of Pt<sup>+</sup> ion fluence, and at a fluence of 10<sup>15</sup> ions cm<sup>-2</sup> the formation of an ordered face-centered tetragonal (FCT) CoPt phase in the mixed region was clearly observed from the grazingincidence x-ray diffraction (GIXRD) measurements. The magneto-optic Kerr effect results show that the coercivity increases with increase in Pt<sup>+</sup> ion fluence.

# 1. Introduction

Synthesis of ordered CoPt materials is receiving a great deal of attention due to their potential magnetic applications [1, 2]. CoPt alloys with near equiatomic composition exhibit face-centered tetragonal (FCT)  $L1_0$  ordered structure. The  $L1_0$  structure is based on a face-centered cubic (FCC) lattice with Co and Pt planes stacked along the [001] direction producing a tetragonal distortion along the *c*-axis. This CoPt phase is a hard magnetic phase with high magnetocrystalline anisotropy where the easy magnetic axis is aligned in the direction of the distorted [001] axis. Tailoring the magnetic properties of these materials is of great interest, and ion irradiation has been shown to be an efficient tool for this [3].

In chemically synthesized CoPt nanoparticles ( $\sim 8$  nm), chemical ordering was observed upon annealing above 600 °C [4], and a similar kind of ordering was also observed in bulk CoPt alloys [5]. The ion-beam mixing of a Co/Pt multilayer film by Ar<sup>+</sup> ion irradiation in an external magnetic field leads to the formation of a chemically ordered CoPt film having a large magnetic moment (2.63  $\mu_B$ ) per Co atom, which is 55% larger than that of the bulk Co (1.73  $\mu_{\rm B}$ ) [6]. These authors reported that the observed gigantic magnetic moment is due to the effect of ultra-fast quenching during the cooling phase of the collision cascade in the presence of an external magnetic field which leads to the formation of CoPt phase with expanded volume. Recently, it has been demonstrated that irradiation may produce chemical order in intermetallic alloys [7, 8]. Significant structural and magnetic ordering was obtained and controlled in FePt films upon He<sup>+</sup> ion irradiation at ~570 K, i.e., well below the ordering temperature. Chappert *et al* [9] have reported that the magnetic anisotropy of Co/Pt multilayers can be changed from perpendicular to in-plane by modifying the Co atom environment near the Pt/Co interface using He ion irradiation. Vieu *et al* [10] modified the magnetic properties of the Pt/Co ultra-thin structure by focused Ga<sup>+</sup> ion irradiation in the energy range 20–100 keV. At lower fluences (<2 × 10<sup>14</sup> ions cm<sup>-2</sup>), the coercive field decreases slowly with irradiation fluence. At higher fluences, collisional intermixing induces significant alloying of the Co layer and the film turns paramagnetic at room temperature. However, Chang *et al* [11] showed that magnetic anisotropy can be induced in a ferromagnetic Co/Pt multilayer film by employing Ar<sup>+</sup> ion irradiation in an externally applied magnetic field.

Most of the works in the literature report experiments done on multilayer films to introduce magnetic anisotropy, and the magnetic order improved in some cases by using He<sup>+</sup> ion irradiation after ion-beam mixing or an external magnetic field during ion-beam mixing. In this paper, for the first time we show that magnetic anisotropy can be induced in a Co/Pt bilayer film by Pt<sup>+</sup> self-ion irradiation at room temperature. Further, we have demonstrated the increase in magnetic anisotropy by high-energy Si<sup>+</sup> ion irradiation at a temperature of 300 °C, which is well below the ordering temperature.

## 2. Experimental details

Using electron-beam evaporation at a rate of 0.1 Å s<sup>-1</sup>, the Pt/Co (400 Å/700 Å) bilayer films with Pt as the top layer were deposited on Si(100) substrate in a ultra-high vacuum chamber with the base pressure of  $8 \times 10^{-10}$  Torr. The samples were irradiated with 600 keV Pt<sup>+</sup> ions with different ion fluences  $(1 \times 10^{15}, 5 \times 10^{15}, 8 \times 10^{15} \text{ ions cm}^{-2})$  at room temperature by using a 1.7 MV Tandetron accelerator. The range of penetration of 600 keV Pt<sup>+</sup> ions in the bilayer sample was found to be 434 Å using the Monte Carlo simulation SRIM 2003 [12]. The sample implanted with 8  $\times 10^{15}$  ions cm<sup>-2</sup> of Pt<sup>+</sup> ions was irradiated with 4 MeV Si<sup>+</sup> ions with the fluence of  $2 \times 10^{16}$  ions cm<sup>-2</sup> while maintaining the sample temperature of 300 °C at a vacuum of  $8 \times 10^{-7}$  Torr. The range of 4 MeV Si<sup>+</sup> ions in the sample is 2.35  $\mu$ m, and hence the implanted Si is deep inside the Si(100) substrate. Rutherford backscattering (RBS) measurements were carried out using 2 MeV alpha particles from the 1.7 MV Tandetron accelerator on as-prepared and ion-irradiated samples under normal incidence and glancing incidence of 70° to increase the depth resolution. The grazing-incidence x-ray diffraction (GIXRD) studies were performed using an x-ray diffractometer (Stoe GmbH, Germany) and a glancing angle of 0.1° was kept for Cu K<sub> $\alpha$ </sub> radiation for all samples. Magneto-optic Kerr effect (MOKE) measurements were performed on the samples with a He-Ne laser. The hysteresis loops were recorded at room temperature by measuring the Kerr rotation with the field applied in the plane of the sample.

## 3. Results and discussion

The RBS spectra of the as-prepared and  $Pt^+$  ion-irradiated samples are shown in figure 1(a). The Co and Pt layers are clearly seen. The composition and thickness of the as-prepared sample were ascertained from the height and width of the spectrum by using GISA3 [13] code. In the



**Figure 1.** (a) The normal-incidence Rutherford backscattering spectra of the as-prepared and Pt<sup>+</sup> ion-irradiated Pt/Co bilayers subjected to a fluence of  $1 \times 10^{15}$  ions cm<sup>-2</sup>,  $5 \times 10^{15}$  ions cm<sup>-2</sup>,  $8 \times 10^{15}$  ions cm<sup>-2</sup>. (b) The glancing incidence (70°) Rutherford backscattering spectra of Pt<sup>+</sup> ion irradiated Pt/Co bilayer subjected to a fluence of  $8 \times 10^{15}$  ions cm<sup>-2</sup> and then irradiated by a 4 MeV Si<sup>+</sup> beam to a fluence of  $2 \times 10^{16}$  ions cm<sup>-2</sup> at 300 °C.

as-prepared sample the platinum and cobalt layer thicknesses were found to be 400 Å and 700 Å respectively. The mixing of Co and Pt across the interface of the Pt/Co bilayer in the irradiated sample is evident from the tailing of the low-energy and high-energy edges of the platinum and cobalt respectively. In the Pt<sup>+</sup> ion-irradiated samples, the reduction in area under the Pt profile and shift in high-energy edges of Si and Co towards higher channels are observed, and these are mainly due to sputtering of the top platinum layer. Figure 1(b) shows the glancing-angle RBS spectra of a Pt<sup>+</sup> ion-irradiated Pt/Co bilayer with a fluence of 8 × 10<sup>15</sup> ions cm<sup>-2</sup> and followed by Si<sup>+</sup> ion irradiation to a fluence of 2 × 10<sup>16</sup> ions cm<sup>-2</sup> at a temperature of 300 °C. The cobalt edge appears at the energy of 1531.4 keV and the silicon edge appears at 940 keV in both spectra. It is evident from both spectra that the step at the cobalt edge at 1531.4 keV is more pronounced after Si<sup>+</sup> ion irradiation. It should be noticed that the increase in the yield of cobalt at energy of 1531.4 keV and the corresponding decrease in the yield of platinum in the Si<sup>+</sup> ion-irradiated sample indicates the alloy formation across the interface.

In order to identify the crystal structure of the alloy of Co and Pt formed across the interface, GIXRD measurements were carried out, and the results are shown in figure 2. The as-prepared sample shows the presence of Pt (FCC) and Co (HCP) phases, as is evident from the diffraction peaks of platinum and cobalt, respectively. The GIXRD pattern of the Pt<sup>+</sup> ion-irradiated sample, subjected to a fluence of  $8 \times 10^{15}$  ions cm<sup>-2</sup>, shows the presence of diffraction peaks of the CoPt phase. Ordering of the CoPt film is evident from the splitting of the (200)<sub>CoPt</sub> and (002)<sub>CoPt</sub> peaks in the GIXRD pattern [4, 5]. The split of the (200)<sub>CoPt</sub> and (002)<sub>CoPt</sub> peaks became more pronounced after Si<sup>+</sup> ion irradiation, indicating enhanced ordering of CoPt phase (refer to the top figure of figure 2). The observed diffraction peaks



**Figure 2.** The GIXRD pattern of the (a) as-prepared, (b) Pt<sup>+</sup> ion-irradiated Pt/Co bilayer subjected to a fluence of  $8 \times 10^{15}$  ions cm<sup>-2</sup> and (c) Pt<sup>+</sup> ion-irradiated Pt/Co bilayer subjected to a fluence of  $8 \times 10^{15}$  ions cm<sup>-2</sup> and then irradiated by a 4 MeV Si<sup>+</sup> beam at a fluence of  $2 \times 10^{16}$  ions cm<sup>-2</sup> at 300 °C. The inset shows the fit for  $(200)_{CoPt}$ ,  $(002)_{CoPt}$  and  $(101)_{Co}$  diffraction peaks of the spectrum given in (c). Inset: the open circles and solid line represent the experimental data points and fitted line respectively. The three curves represented by dashed lines are the Voigt profiles selected in the peak fit code. The bottom straight dashed line is the background.

(This figure is in colour only in the electronic version)

of platinum are due to unmixed platinum present in the sample. The cobalt peaks are more prominent in the implanted sample in comparison to the as-prepared sample. This effect could be ascribed to the fact that, in the as-prepared sample, the Co is buried by Pt and not seen by the x-rays under grazing incidence but it becomes visible when most of the Pt has been sputtered away by irradiation. The diffraction peaks of platinum in the GIXRD pattern vanished after  $Si^+$ ion irradiation at 300 °C, indicating complete mixing of platinum and cobalt. Chang et al [6] has reported a shift of  $0.4^{\circ}$  of the (111) peak position of the CoPt phase to lower angles in Ar<sup>+</sup> ionirradiated Co/Pt multilayers, and have attributed it to expanded lattice spacing. In the present experiments, a shift of 0.36° of the (111) peak position of CoPt phase from standard JCPDS data was observed in the Pt<sup>+</sup> ion-irradiated Pt/Co bilayers that were followed by high-energy Si<sup>+</sup> ion irradiation at elevated temperatures. The unit cell dimensions a, c for the Si<sup>+</sup> ion-irradiated sample were calculated to be 3.869 Å and 3.767 Å from the peak positions of the (200)<sub>CoPt</sub> peak  $(2\theta = 46.9^\circ)$  and  $(002)_{CoPt}$  peak  $(2\theta = 48.26^\circ)$ . The c/a ratio is found to be 0.974, which is in close agreement with one of the values obtained for the FCT CoPt phase [14]. In the present experiments the formation of ordered CoPt phase is seen directly upon Pt<sup>+</sup> ion irradiation on Co/Pt bilayers itself, unlike the CoPt<sub>3</sub> and CoPt phase (ordered/disordered) formation upon Kr<sup>+</sup> ion irradiation on Co/Pt bilayers [15].

For CoPt alloy the critical temperature  $(T_c)$  for the radiation-enhanced diffusion (RED) process is calculated by using the empirical relation  $T_c = 95.2\Delta H_{coh}$  (eV/atom) [16, 17], and found to be 214 °C. The Si<sup>+</sup> ion irradiation was carried out at a temperature of 300 °C,



Figure 3. MOKE signals of Pt<sup>+</sup> ion-irradiated Pt/Co bilayers subjected to a fluence of 1 ×  $10^{15}$  ions cm<sup>-2</sup>, 5 ×  $10^{15}$  ions cm<sup>-2</sup>, 8 ×  $10^{15}$  ions cm<sup>-2</sup>. The MOKE signal of Pt/Co bilayer irradiated with Pt<sup>+</sup> ions at a fluence of 8 ×  $10^{15}$  ions cm<sup>-2</sup> and then irradiated by a 4 MeV Si<sup>+</sup> beam at a fluence of 2 ×  $10^{16}$  ions cm<sup>-2</sup> at 300 °C is also shown.

Table 1. The coercivity values of ion-irradiated samples.

Fluence	Coercivity (Oe)
$1 \times 10^{15}$ ions cm <sup>-2</sup> of Pt	141.5
$5 \times 10^{15}$ ions cm <sup>-2</sup> of Pt	417.2
$8 \times 10^{15}$ ions cm <sup>-2</sup> of Pt	655.8
$8 \times 10^{15}$ ions cm <sup>-2</sup> of Pt and	776.7
$2 \times 10^{16}$ ions cm <sup>-2</sup> of Si	

which is higher than the  $T_c$  for RED to occur. Kinetic Monte Carlo simulations by Bernas *et al* [18] show that the following requirements have to be met by the irradiating beam to initiate ordering in the target material: (a) small energy transfers, (b) minimizing recoil displacements, and (c) a low collision cross section in order to avoid defect interactions. As per the SRIM calculation [12], the energy absorbed by the recoils in CoPt film from 4 MeV Si<sup>+</sup> ions is 54.03 eV/incoming ion/nm, and the number of displaced atoms is  $60.35 \times 10^{-2}$ /incoming ion/nm. Because of the low-energy transfers and small number of displaced atoms produced by 4 MeV Si<sup>+</sup> ion irradiation, chemical ordering of CoPt phase is favored.

The ordering of CoPt is expected to increase the magnetocrystalline anisotropy. In order to observe the increase in magnetocrystalline anisotropy, MOKE measurements were carried out on the as-prepared and ion-irradiated samples. The MOKE signals of the ion-irradiated samples are shown in figure 3. The as-prepared sample does not show any hysteresis because the top Pt layer attenuates the light. The coercivity values for Pt<sup>+</sup> and Si<sup>+</sup> ion-irradiated samples are given in table 1. The hysteresis loop of the Pt<sup>+</sup> ion-irradiated sample subjected to the fluence of  $1 \times 10^{15}$  ions cm<sup>-2</sup> of Pt<sup>+</sup> ions exhibits a square shape, which indicates a large amount of

unmixed cobalt. It is observed that the coercivity increases as a function of irradiation fluence, indicating the increase in magnetocrystalline anisotropy.

In summary, we have studied the changes in properties of self-ion-implanted Pt/Co film, followed by high-energy  $Si^+$  ion irradiation at elevated temperatures. We have observed the interface mixing and the formation of ordered CoPt phase directly, by Pt<sup>+</sup> ion irradiation. Further ordering of the CoPt phase was observed by high-energy Si<sup>+</sup> ion irradiation at elevated temperatures. MOKE measurements indicate the increase in magnetocrystalline anisotropy upon Pt<sup>+</sup> ion irradiation.

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