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Pt diffusion driven $L1_0$ ordering in off-stoichiometric FePt thin films

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ABSTRACT

Pt/Fe_3Pt thin films were deposited on the ⟨100⟩ silicon substrates at room temperature using DC sputtering. As-deposited Pt (25 nm)/Fe_3Pt (70 nm) bilayers were subjected to heat treatment and a systematic study was carried out to investigate the structural and magnetic phase transition as a function of annealing temperature. The as-obtained films show an A1 disordered, face centered cubic phase which exhibit a magnetically soft behavior. After annealing at 300 °C for 1 h, the set in of $L1_0$ face centered tetragonal phase was observed and is attributed to the atomic diffusion at Pt/Fe_3Pt interface. A coercivity of 6.8 kOe was obtained for Pt/Fe_3Pt at 300 °C which increases to 12.5 kOe at 400 °C due to enhanced fct phase formation. Rutherford back scattering study has also been performed to investigate the inter-layer diffusion of Fe_3Pt and Pt, along with estimation of film thickness and atomic composition with annealing temperature. The thickness and composition of films annealed at 600 °C was found to be 73 nm and $Fe_{54}Pt_{46}$, respectively. Lower temperature ordering is achieved for these bilayer films.

1. Introduction

FePt and CoPt nanostructured materials have become an extensive area of research interest due to their wide applications in biomedical diagnosis [1,2] and ultra-high density magnetic recording [3,4]. Both FePt and CoPt in $L1_0$ phase possess hard magnetic behavior with high magnetocrystalline anisotropy (K_u) [5,6] which makes the material more suitable for the recording applications with small grain size. A lot of research is focused on the fabrication process of $L1_0$ ordered FePt thin films. Several methods of film deposition are reported including chemical methods [3,7], sputtering [8,9], pulsed laser deposition [10,11], molecular beam epitaxy [12,13] and electrochemical deposition [14]. Sputtering is one of the most well-known techniques used for multi-layered thin film deposition. It gives a very good control on the thickness, uniformity and composition of thin films.

FePt exhibits order–disorder transformation from A1 (disordered) phase to $L1_0$ (ordered) phase. This phase transformation is very much dependent on the substrates, underlayers and deposition conditions [15]. As-deposited thin films possess disordered face centered cubic (fcc) structure which can be transformed to ordered face centered tetragonal (fct) structure either by carrying out the in situ heat treatment during film deposition [16,17] or by post annealing treatment at different temperatures [18–20]. Deposition at higher temperature or post annealing lead to grain growth [21] which is not desirable for these materials to be used for practical applications. So, the need is to obtain smaller size particles along with low ordering temperature. Several reports are available in literature in which the ordering temperature is reduced by modifying the surface with different metal oxides or metal layer [22–25], co-depositing the equiatomic FePt along with different metals [26,27], ion irradiation [28,29], plasma focus ion irradiation [30,31], depositing multilayers of Fe/Pt [32,33] etc.

In many of the earlier reports, different metal underlayers have been used to enhance the structural and magnetic properties. The interlayer diffusion of these metal underlayers that take place at the metal–FePt interface during annealing may lead to formation of metal alloys other than FePt [34,35]. To avoid this situation, in this report, a new and significant way of reducing the ordering temperature has been investigated. We have used Pt deficient FePt target for thin film fabrication and the composition of the FePt alloy is adjusted by the addition of Pt underlayer. The Pt underlayer is used for achieving the ordered phase at lower temperature. Pt not only serves as a source for the composition gradient but also reduces the compressive stress which is essential for the formation of $L1_0$ phase. The formation of ordered structure is investigated as a function of annealing temperature (300–600 °C). We observed that the coercivity increases with annealing temperature and is strongly correlated with ordering parameter. The reduction in the ordering temperature has been demonstrated successfully.

2. Experimental

FePt thin films were fabricated in a high vacuum DC sputtering system (HINDHIVAC, model 12″ MSPT) using Fe rich Fe_3Pt alloy

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with Fe:Pt off stoichiometric atomic composition of around 73:27 (estimated using RBS) and Pt target (99.99% purity). Pt/Fe_3Pt bilayers were deposited at room temperature on single crystal Si substrates oriented along ⟨100⟩. High purity argon at a pressure of 0.03 mbar was introduced into the sputtering chamber, having base pressure of 2×10^{-6} mbar, to get the Pt and Fe_3Pt thin films. The deposition was carried out at 300 V and 0.1 Amp current. The time of deposition of Pt and Fe_3Pt was optimized to 1 min and 3 min respectively. The thickness of Pt and Fe_3Pt were estimated to be 25 nm and 70 nm respectively, using Rutherford back scattering (detailed analysis is discussed in Section 3.4). The deposited films were subjected to annealing in Ar+H_2 atmosphere in the temperature range 300–600 °C for 1 h.

The crystallographic studies were carried out using X-ray diffractometer (Discover D-8) equipped with Cu-Kα radiation of $\lambda=1.5405$ Å, operated at 40 kV, 40 mA. Magnetic properties of the films were examined using vibrating sample magnetometer (EV-9, Microsense) with a maximum applied magnetic field of ±22 kOe. The thickness and composition of the alloy films were estimated using Rutherford back scattering (RBS) performed with 2 MeV α particle beam at a scattering angle of 170°. The surface morphology were analyzed by field emission scanning electron microscopy (FE-SEM, Mira II LMH, Tescan).

3. Results and discussion

3.1. Structural studies

Fig. 1 shows the XRD pattern of Pt/Fe_3Pt in as-deposited state and annealed at different temperatures in the range 300–600 °C for 1 h. As-deposited film (Fig. 1a) shows the peaks at 39.98° and 46.59° corresponding to (111) and (200) planes respectively of Pt and the weak and broad shoulder present at 40.89° corresponding to the fundamental (111) plane indicating that the as-deposited Pt/Fe_3Pt films exhibit face centered cubic (fcc) disordered phase. After annealing at 300 °C, the fundamental peaks of FePt are observed at 40.95° and 47.0° corresponding to the (111) and (200) planes respectively (Fig. 1b). Along with this, Fe_3Pt is also present (marked as*) at the lower angle side of (111) peak of FePt. The superlattice peaks at 23.82° and 32.82° starts appearing, confirming the initiation of ordering of FePt. The observed shift in the fundamental peak after annealing at 300 °C is due to the atomic rearrangement of Fe and Pt in the partially ordered FePt unit cell. The bigger atomic radii of Pt as compared to Fe may induce the asymmetry in the unit cell leading to the expansion in lattice parameter '*a*' and reduction in lattice parameter '*c*'. This indicates the evolution of ordered phase at 300 °C which can be

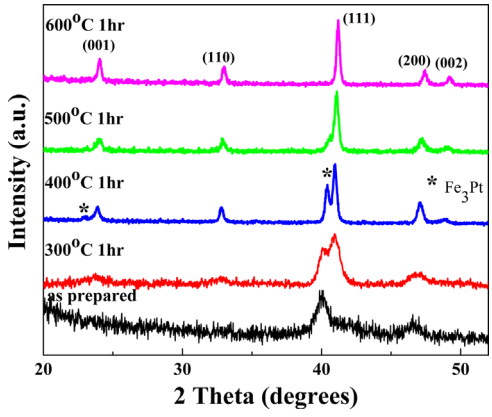

Fig. 1. XRD pattern of Pt/Fe_3Pt films (a) as-deposited, annealed for 1 h at (b) 300 °C (c) 400 °C (d) 500 °C and (e) 600 °C.

further enhanced by annealing the specimen at 400 °C. At 400 °C, the superlattice peaks appear clearly and splitting of peaks corresponding to (200) and (002) have also been observed confirming the transformation to face centered tetragonal (fct) phase from fcc phase as shown in the Fig. 1c. However, peaks corresponding to (111) Fe_3Pt and (111) FePt becomes sharper due to the increase in crystallinity and grain growth. There is a possibility that the FePt and Pt interface has been transformed partially to $L1_0$ FePt phase and the rest is Fe_3Pt due to diffusion of Pt at the interface. The Pt diffusion from Pt underlayer to Fe_3Pt layer and vice versa takes place and the rate of diffusion increases with increase in annealing temperature. The Fe_3Pt peaks are getting suppressed with increase in annealing temperature and it disappears completely at 600 °C resulting into the transformation to an ordered $L1_0$ FePt phase (Fig.1d,e). With an increase in annealing temperature, the (001) peak position shifts to higher angle indicating the contraction of unit cell along *c* axis while the (110) peak position also shifts to higher angle which may be due to the presence of Fe rich Fe_3Pt phase along with $L1_0$ phase. This may be due to the fact that d_{110} of Fe_3Pt is slightly greater than d_{110} of FePt. The decrease in the full width at half maxima (FWHM) with annealing shows the increase in crystallite size. The average crystallite size estimated using the Scherrer formula for the as deposited FePt is 4 nm which has increased to 37 nm after 600 °C annealing.

The presence of the Pt underlayer induces a stress which is found to be released on annealing [25]. However, this reduced compressive stress is found to decrease the ordering temperature. A quantitative estimate of the compressive stress has been performed to understand this behavior. The compressive stress (σ) can be estimated using the following relation:

$$\sigma=\Delta\alpha\Delta T\left(\frac{E}{(1-\upsilon)}\right) \quad (1)$$

where $\Delta\alpha$ is the difference between coefficients of thermal expansion, ΔT is the difference between room temperature and annealing temperature, E is the elastic modulus of the film and υ is the Poisson ratio [15]. The compressive stress at 300 °C annealing temperature with and without Pt underlayer was estimated to be 181 MPa and 573 MPa respectively. A reduction in σ is very clear. Also, the presence of compressive stress indicates that the *c* axis lies preferentially in the plane of film. In addition to the compressive stress, the diffusion of Pt also promotes the ordering. Hsu et al. demonstrated such a behavior on FePt deposited on Pt coated quartz substrates [25].

The chemical ordering is characterized by the ratio of integrated peak intensity of (001) and (002) i.e. I_{001}/I_{002}. The ratio of I_{001}/I_{002} increases with annealing temperature and reaches a value of 3.16 at 600 °C suggesting an increase in long range ordering on heat treatment. The ordering parameter, *S* calculated using lattice parameters '*a*' and '*c*' is 0.941 for the 600 °C annealed sample. The ratio of I_{001}/I_{111} is a measure of *c*-axis orientation. The ratios of I_{001}/I_{111} for the 300 °C and 600 °C annealed samples are 0.16 and 0.40 respectively indicating that the (001) orientation is also improved with annealing. Table 1 summarizes the above results.

3.2. Magnetic studies

Magnetic measurements performed in the in plane and out of plane geometry on the as-deposited films and films annealed at different temperatures are shown in Fig. 2. The as-prepared films (Fig. 2a) are magnetically soft in behavior confirming the disordered A1 phase which is in confirmity with the XRD analysis. The hysteresis loop obtained on the sample annealed at 300 °C for 1 h shows a marked increase in coercivity, H_c~6.76 kOe as shown in Fig. 2b which further enhances to 12.52 kOe at 400 °C (fig. 2c).