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Effect of Nitrogen on Magnetic Properties of FePt-Al_2O_3 Granular Thin Film

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The influence of nitrogen on the magnetic and crystallographic properties of sputtered, FePt-Al_2O_3 films with 20 nm thickness have been studied. The total gas pressure (P_N+P_{Ar}) was set at 5 mTorr. This paper revealed that the proper amount of nitrogen led to decrease the ordering temperature, and enhance the coercivity as well as make the crystallite size smaller. The saturation magnetization decreased with increasing amount of nitrogen, but the coercivity increased up to a certain limit then decreased. Maximum coercivity of 7.5 kOe was found for the film deposited at P_N of 0.75 mTorr followed by annealing at 500°C for 60 minute, whereas it was about 0.75 kOe for the film without nitrogen annealed in same the condition. The minimum average crystallite size, evaluated with x-ray diffraction line, was found to be about 5 nm for the film prepared at P_N of 0.75 mTorr. γ'-Fe_4N phase was observed along with FePt even at low P_N of 0.25 mTorr only.

Key words: nitrogen, FePt-Al_2O_3, thin film, magnetic recording.

1. INTRODUCTION

The popularity of the computer internet and mobile digital recording devices has further increased the demand of high recoding density for the storage media. Focused on increasing the areal density of magnetic recording, much effort has long been done [1,2]. In order to increase storage density, the volume of each bit cell should be reduced dramatically but for any given materials, there is a critical grain size where thermal fluctuation becomes dominant at room temperature. The thermally unstable media can loose stored data easily. The product of the anisotropy energy (Ku) and the volume of the magnetic switching unit (V) represent the energy barrier to magnetization reversal in the presence of thermal fluctuations. If $KuV/k_bT > 60$, the magnetic medium is considered thermally stable (T : temperature and k_b: Boltzmann constant) [3]. Hence, it is essential to search for new materials that have higher anisotropy energy to continue to increase the magnetic recording density. FePt films with $L1_0$ ordered (FCT) structure is a prominent candidate for ultrahigh-density recording media due to its high magnetocrystalline anisotropy, high thermal stability, and good chemical stability [4]. By itself , FePt is not a viable media material because the requisite processing temperatures result in large grains (40~60 nm) that are highly exchange coupled. Additional agents are needed to tailor the thin-film microstructures and magnetic properties. Elements such as Al_2O_3, MgO, TaN, Ag, [5]- [8], have shown promise in controlling FePt grain size in composite films. Other studies have shown that using B [9], BN [10] additives can reduce the magnetic coupling and Cu [11], Zr [12] enhance the ordering in FePt thin films. It is worthy to note that, in our previous study [13] we have achieved an excellent fine grain about 8-10 nm and low surface roughness with a film thickness of only 20 nm by using Al_2O_3 matrix with FePt film but the ordering temperature was as high as 550°C and coercivity was not so good.

Insertion of nitrogen in FePt during sputtering without substrate heating may produce following effects, 1) The pushing effect of interstitial nitrogen can break the large crystallite into smaller one. 2) During annealing, N_2 may leave the crystal and produce some free spaces that can enhance the ordering process. 3) Chemical bonding of Fe and N atoms through electron donation from N atoms to incomplete d sublevels of Fe. By varying the nitrogen atoms in the FePt-Al_2O_3 films it may be possible to tailor the magnetic as well as crystallographic properties.

In this article, we have fabricated the FePt-Al_2O_3 thin films at various partial pressure of N_2 (P_N) followed by annealing in vacuum at different temperatures and their magnetic properties and crystallographic characteristics were investigated.

2. EXPERIMENTAL PROCEDURE

The $Fe_{52}Pt_{48}$-Al_2O_3 (25 vol. %) thin films with 20nm thickness were prepared by using RF magnetron sputtering system on glass (Corning 7059) substrate without substrate heating. After evacuating the chamber to a pressure below 2×10^{-6}Torr, argon gas (99.99% in purity) and nitrogen gas (99.99% in purity) were introduced. The discharge gas pressure (P_{total}), which is the sum of each partial pressure (P_{Ar}, P_N), was kept constant at 5mTorr. The partial pressure of nitrogen was varied from 0 – 2.5mTorr. The sputtering power was 80W. A mosaic target consisting of alumina disk overlaid with iron plate and platinum chips was used. Post deposition annealing of all films were done at various temperatures from 300°C to 500°C for one hour in vacuum better than 5×10^{-6} Torr to obtain magnetically hard FCT structure.

Film compositions were determined by electron probe x-ray micro analysis (EPMA). Crystallographic properties of the films were investigated by an X-ray diffractometer (XRD). A vibrating sample magnetometer (VSM) with a maximum field of 15kOe was used to measure the magnetic properties of the thin films.

3. RESULTS AND DISCUSSION

Fig.1 shows the Magnetization curves for FePt-Al_2O_3 films with and without N_2 after post deposition annealing at various temperatures for one hour. In as deposited state, the magnetization curves for both kind of films are magnetically soft because of its randomly oriented FCC structure. Coercivity of the FePt-Al_2O_3 films deposited at P_N of 0 mTorr and 0.75mTorr start to increase after annealing at 500 ℃ and 400 ℃ respectively, indicating the beginning of transformation to FCT $L1_0$ ordered structure[14] . In case of FePt-Al_2O_3 films deposited at P_N of 0.75 mTorr, a drastic increase in coercivity occurs at 450℃, whereas similar effect has been found for the FePt-Al_2O_3 film without N_2 after annealing at temperature higher than 550℃[13]. This shows

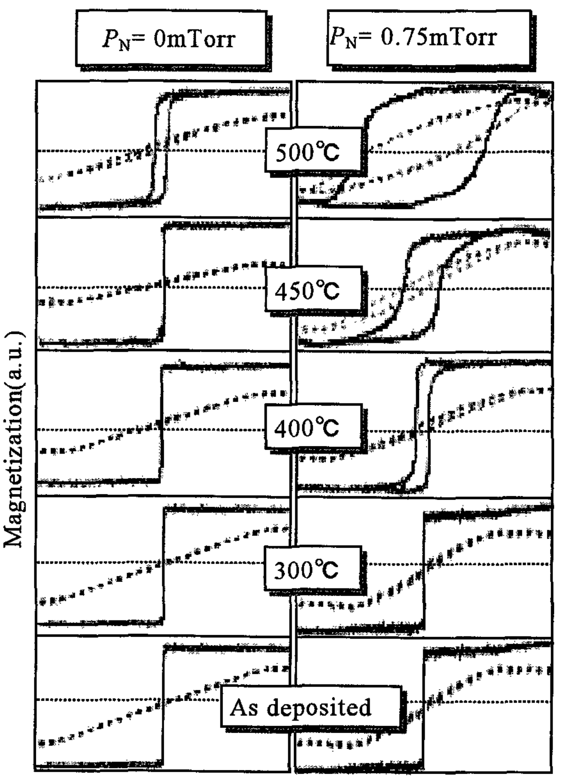

Fig.1. Magnetization curves for FePt-Al_2O_3 films without (left) and with (right) nitrogen after ex-situ annealing at various temperatures for one hour. The magnetic field was applied in the in-plane direction (solid line) and in the perpendicular direction (broken line).

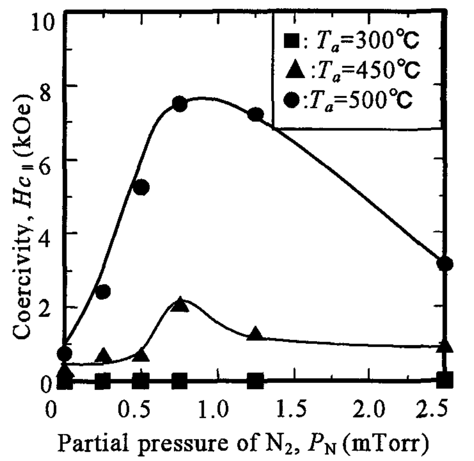

Fig.2. Plot of in-plane coercivity vs. partial pressure of N_2 (P_N) for FePt-Al_2O_3films annealed at different temperatures for one hour.

that the addition of N_2 to the FePt-Al_2O_3 film is very effective in reducing the temperature for ordering. The demagnetization curve of the FePt-Al_2O_3 films prepared with N_2 show an inflection point, this may be due to the presence of γ'-Fe_4N phase.

Fig.2 shows the variation of in-plane coercivity of the FePt-Al_2O_3 films with P_N annealed at different temperatures for one hour. The coercivity of the films annealed at 300℃ is zero for the any value of P_N because the films are in disordered state. In case of films annealed at 450℃, the coercivity increases from zero with the increase in P_N to a maximum value of about 2.0 kOe at P_N of 0.75 mTorr then decreases to about 1.0 kOe and remains almost constant for higher values of P_N. Similar trend is observed for the films annealed at 500℃ but the coercivity value is much higher and it decreases sharply at higher values of P_N . The maximum coercivity is about 7.5 kOe obtained at P_N of 0.75 mTorr whereas it is about 0.75 kOe for the film without N_2. The increase of coercivity value with the P_N is not so clear, it may be due to an expansion of lattice volume by interstitial N_2, that may alter the exchange interaction and thereby increase the coercivity [15].

The effect of nitrogen partial pressure on saturation magnetization is shown in Fig.3. It can be easily seen from the figure that, the saturation magnetization decreases drastically with the increase of P_N up to about 0.75 mTorr then it remains almost constant for the higher values of P_N. The trend is similar for the samples annealed in 450℃and 500℃. In the FePt system the magnetization process mainly depends on the individual magnetic moments borne by the iron atoms and their exchange interactions. The bonding between the iron atoms and nitrogen can be regarded as interstitial where the intermixing of 3*d* orbital of Fe and 2*p* orbital of N are energetically favorable. These kinds of interactions can be understood from the idea of fractional bonds introduced by Paulings [16]. There are enough evidences [17,18], which describes the electron donation ability of the nitrogen atoms to the incompletely filled 3*d* or more

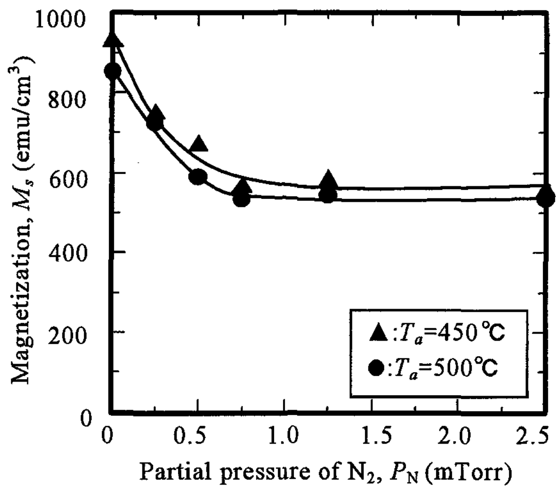

Fig.3. Plot of saturation magnetization (Ms) vs partial pressure of N_2 (P_N) for FePt-Al_2O_3 films annealed at 450°Cand 500°C for one hour.

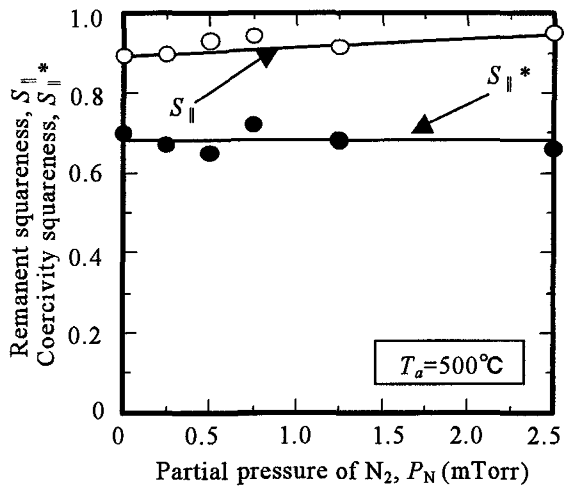

Fig.4. Plot of in-plane squareness (S) and coercivity squareness (S*) vs partial pressure of N_2 (P_N) for FePt-Al_2O_3 films annealed at 500°Cfor one hour.

correctly *spd* band of Fe in several materials with the nitrogen as interstitials. The electron supplying nitrogen atoms may populate the spin down states of Fe, and result in a reduction of the intrinsic magnetic moment of Fe atoms.

Fig.4 shows the influence of P_N on the remanent squareness S and coercivity squareness S*. The squareness S slightly increases with the increasing values of P_N. The trend of coercivity squareness, S* does not show prominent changes upon nitrogen addition, it remains almost constant at 0.7.

XRD diagrams of FePt-Al_2O_3 and FePt-Al_2O_3-N films annealed at different temperatures for 1 hour is shown in Fig.5 (a) and (b) respectively. The as deposited state of both films are disordered FCC. In the case of FePt-Al_2O_3 film without N_2, a FePt (111) $_{FCC}$ peak is found around 40° for 2θ in the as deposited state which diminishes with the increase of annealing temperature and start to appear FePt (111) $_{FCT}$ peak around 41.2 ° for 2θ. The films deposited in P_N of 0.75mTorr do not show any peak up to 300°C which indicates that N_2 suppress the formation FCC FePt (111) texture. Upon annealing it begins to show fundamental FePt $(111)_{FCT}$ and another peak of γ'-$Fe_4N(200)_{FCC}$, with increasing annealing temperature the intensity of these peaks increase. The effect of the ferro and soft magnetic γ'-Fe_4N on the magnetic properties of FePt-Al_2O_3 film is not well understood in this time. The intensity of FePt $(111)_{FCT}$peak in FePt-Al_2O_3-N film is smaller than

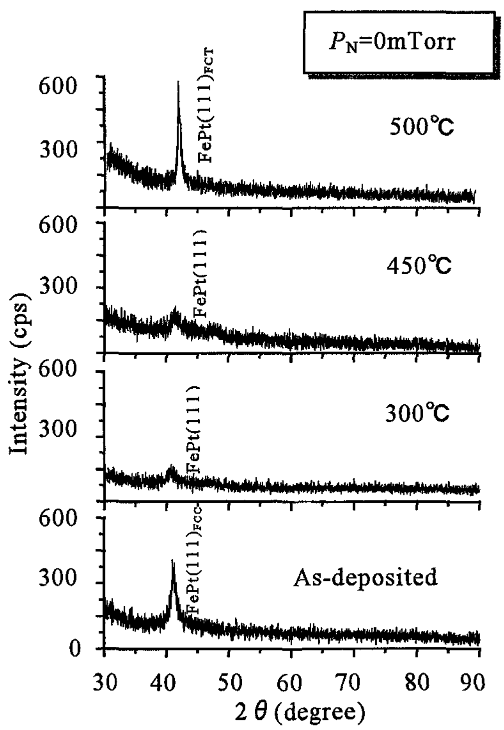

Fig.5(a). XRD diagrams of FePt-Al_2O_3 films annealed at different temperatures for one hour.

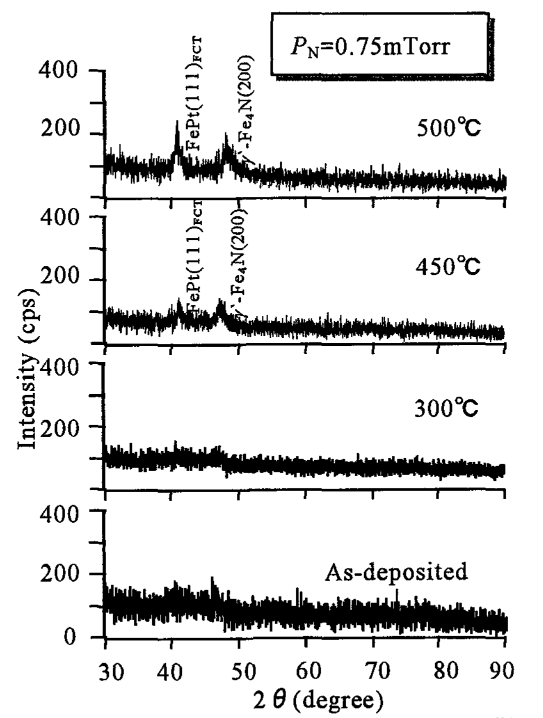

Fig.5(b). XRD diagram of FePt-Al_2O_3-N films annealed at different temperatures for one hour.

that of FePt- Al_2O_3 films but due the smaller grain size, the coercivity is much higher in former case. The drastic increase in coercivity for the films deposited at P_N of 0.75 mTorr and annealed at 450℃ indicates the formation of $L1_0$ structure at this temperature but we have not found any super lattice peak in the XRD pattern even annealed at 500℃. This may be due to the low thickness of the films. The full width at half maximum of FePt (111) main peak in XRD diagrams increases with the P_N indicating a decreasing crystallite size of the films.

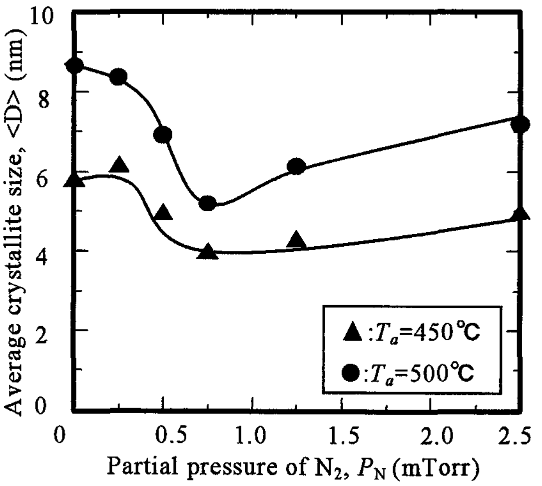

Fig.6. Plot of XRD crystallite size vs partial pressure of N_2 (P_N) for FePt-Al_2O_3 films annealed at 450℃and 500℃for one hour.

Fig.6 shows the dependence of FePt crystallite size with the variation of partial pressure of N_2. The average crystallite size <D> estimated from the FePt (111) peak width of the XRD diagrams using Scherrer's formula, decreases with the increasing of P_N up to 0.75 mTorr then increases with the increasing amount of N_2. Iron and its alloy have remarkable affinity towards nitrogen atoms, this is due to the intermixing of N-2*p* sates with Fe-3*d* states because of similarities between their density of states [19]. The pushing effects of atomic nitrogen into the FePt crystal may result in breakage of larger crystallites into smaller ones. The rate of grain growth with the temperature is also suppressed by the addition of N_2.

4. CONCLUSION

$Fe_{52}Pt_{48}$-Al_2O_3 (25 vol.%) films were fabricated in N_2 (0-2.5 mTorr) plus Ar atmosphere by RF magnetron sputtering system. Magnetic and crystallographic properties of these films were characterized after annealing in vacuum at different temperatures for one hour. The magnetic and crystallographic properties as well as ordering characteristics of FePt- Al_2O_3 granular thin films are improved remarkably due to the addition of limited amount of N_2 with Ar gas during sputtering.

The most significant improvement was observed in the films prepared at P_N of 0.75 mTorr. The highest coercivity of about 7.5 kOe for the films annealed at 500℃ for one hour and the lowest ordering temperature of 450℃ were observed in this condition. Further understanding of the interdependence of P_N, stoichiometry, and microstructure of FePt-Al_2O_3 granular thin film is needed.

5. REFERENCES

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