Reduction in L1₀ Phase Transition Temperature of PLD Grown FePt Thin

Films by Pre-annealing Pulsed Laser Exposure

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Abstract

A pre-annealing atmospheric pulsed laser exposure was applied to decrease the phase transition (from chemically disordered A1 phase to chemically ordered $L1_0$ phase) temperature of FePt nano-particles on a Si (100) substrate. Different pre-annealing laser energy densities of 0.024 and 0.079 J/cm² were utilized to expose the pulsed laser deposition (PLD) FePt thin film samples under atmospheric conditions. Subsequently, FePt thin film samples were annealed at different temperatures of 300 and 400 °C to observe the influence of laser exposure on the phase transition temperature. The phase transition temperature was decreased from conventional 600 °C to 400 °C by one shot pre-annealing atmospheric pulsed laser exposure.

1. Introduction

L1₀ ordered equiatomic FePt thin films are a strong candidate for ultrahigh density magnetic recording media, since its large anisotropy constant K_u (6.6×10^6 to 1×10^7 J/m³) allows a thermally stable small grain size of about 4 nm, which is expected to achieve the storage density greater than 1 Tb/in² [1, 2]. However, as-deposited FePt nanoparticles are normally in chemically disordered A1 face-centered-cubic (fcc) phase and magnetically soft. Post-annealing is required for the phase transformation from fcc to $L1_0$ ordered face-centered-tetragonal (fct) phase which shows ferromagnetic properties. The conventional transition temperature to magnetically hard L1₀ phase, reported in literature, was greater than 550 to 600 °C [3, 4], which causes grain growth and agglomeration of magnetic nanoparticles. Therefore, to achieve an ultra-high areal density of data storage, lower L1₀ phase transition temperature is required. Several approaches have been attempted to lower the transition temperature, such as ion irradiation [2, 5-8], incorporation of a third element [9], magnetic trapping assisted PLD [10] and pulsed laser annealing [11, 12]. Pulsed laser annealing allows rapid heating of thin films in an extremely short period of time (100 fs-100 ns), which prevents heat diffusion. Saita and Maenosono [11, 12] and Inaba et al [11, 12] found that phase transition from fcc phase to fct phase can be directly obtained in FePt thin films by using pulsed laser annealing without any thermal annealing. However, only longitude orientation in FePt thin films was achieved [11, 12]. In this paper, perpendicular orientation of FePt nanoparticles in fct phase was obtained by using pre-annealing one shot pulsed laser exposure and thermal annealing. Furthermore, the phase transition occurs at about 400 °C,

which is about 150 to 200 °C lower than the normally required thermal annealing temperature (550 to 600 °C).

2. Experiments

PLD has been employed for the synthesis of FePt thin films in vacuum (with an operating pressure better than 5×10^{-6} mbar) at room temperature. A continuum Nd: YAG laser (532nm, 10 Hz, 10 ns, 26 mJ and 42.71 J/cm²) was focused on FePt (50:50 at%, 99.99%) target disc to ablate the material which was deposited on rotating Si substrates placed in front of it. After FePt thin films deposition by PLD, pre-annealing pulsed laser exposure was carried out using Continuum Surelite II-10 Nd:YAG laser (532nm, 10Hz, 1 shot) at different energy densities of 0.024 and 0.079 J/cm². The detailed experimental setup for pulsed laser exposure is shown in *Figure 1*. Lens1 and Lens2 were employed to expand the beam size in order to decrease the laser energy density to a value lower than the ablation threshold (1 to 2 J/cm²) [13] of FePt and also to cover the entire surface of the sample being exposed. An aperture was applied to select a more uniform part of the laser beam for exposure.



Figure 1. Schematic diagram of pre-annealing laser exposure setup

The thin film samples were exposed to only one laser shot at the given laser areal energy density. The laser exposed samples were then annealed to different temperatures of 300 and 400 °C and maintained at that temperature for 1 h in atmosphere before cooling down naturally.

The thickness $(96 \pm 3 \text{ nm})$ and composition (44.5 at% Fe and 55.5 at% Pt) of as-deposited FePt thin films were estimated from JOEL JSM-6700F Field Emission Scanning Electron Microscope (FESEM) coupled with Oxford Instrument's Energy Dispersive X-ray (EDX) spectrometer. The structural properties of thin films were analyzed by using SIEMENS D

5005 Cu K_a (1.5406 Å) X-ray diffractometer (XRD). Magneto-optical Kerr effect (MOKE) magnetometer was applied to analyze the magnetic properties of thin films.

3. Results and Discussion

The X-ray diffraction (XRD) patterns are shown in *Figure* 2 to investigate the effect of pre-annealing laser exposure of different energy densities on the phase transition temperature.



Figure 2 XRD patterns of samples exposure at various laser energy densities and annealed at various temperatures of 300 and 400 °C.

TABLE I X-ray peak intensities and crystallite size for FePt without laser exposure and with laser exposure with energy densities of 0.024 and 0.079 J/cm² at annealing temperature of 400 °C.

Laser	Exposure	Energy	$I_{(001)}/I_{(111)}$	$I_{(001)}/I_{(002)}$	Crystallite Size
Density	(J/cm^2)				(nm)
Without Laser Exposure			0	0	9.84
0.024			0.501	2.05	10.66
0.079			0.584	1.04	7.70

The samples, either with or without laser exposure, exhibit the fcc phase with a single broad peak of (111) at about 41° after post-annealing at 300 °C. When the annealing temperature was increased to 400 °C, more peaks, like (001) and (002), appear on the XRD patterns of the samples with laser exposure in comparison to the sample without laser exposure, indicating a lower annealing temperature of the phase transition from low K_u fcc phase to high K_u fct phase. The appearance of (001) peak indicates that the FePt nanoparticles have been

converted to chemically ordered fct phase, because the intensity ratio, $I_{(001)}/I_{(111)}$, is larger than 0.5 for the fct phase FePt but zero for the fcc phase FePt (refer to TABLE I) as the (001) diffraction peak is not observed. The order parameter, which is calculated from the ratio of the x-ray peak intensities of (001) and (002) peaks, ranges from 0 for the chemically disordered fcc phase to 1 for the fully ordered L1₀ fct phase and can be used to measure the long range L1₀ order. The estimated order parameters with laser exposure also indicates that laser exposure induces the phase transition from fcc to fct at a lower temperature (refer to TABLE I). Moreover, the average crystallite size of FePt, calculated by the Scherrer formula, is estimated and shown in TABLE I. The small difference between the average crystallite size of the laser exposure samples and that of without laser exposure samples indicates that laser exposure has small influence on grain growth, which is useful for its application in ultrahigh data storage. Thus, XRD patterns indicate a decreased phase transition temperature for FePt due to pre-annealing pulsed laser exposure.



Figure 3 Representative in-plane hysteresis loops of as-deposited (solid line) and pre-annealing pulsed laser exposed (dash line: 0.024 J/cm²) samples without post-annealing.

Figure 3 shows the representative in-plane hysteresis loops, obtained using MOKE magnetometer, of as-deposited and pre-annealing pulsed laser exposed (0.024 J/cm^2 , 1 shot) FePt thin films. It can be seen that the coercivity of the sample increased from 13.32 kA/m to 40.00 kA/m after one shot laser exposure. The hysteresis loops of FePt thin films exposed with energy density of 0.079 J/cm² also showed an increased coercivity in comparison to

as-deposited FePt thin films. The increased coercivity is due to the phase transition from low K_u fcc phase to high K_u fct phase. This indicates that pulsed laser exposure improves the hard magnetic characteristics of FePt thin films and decreases the post-annealing phase transition temperature.

4. Conclusion

A pre-annealing pulsed laser exposure process is applied on FePt thin films, synthesized by PLD, with an aim to lower the phase transition temperature from soft (fcc) to hard (fct) magnetic phase. It was observed that pre-annealing laser exposure can lower down the annealing temperature from about 600 °C to 400 °C to induce $L1_0$ ordered fct phase transition from chemically disordered A1 fcc phase of FePt thin films. The pre-annealing laser exposure has been found to improve the magnetic properties of FePt thin films, indicating its possible application in ultrahigh data storage. The positive influence of pre-annealing laser exposure on phase transition temperature and magnetic properties of FePt thin films also indicates the possible potential applications of laser interference to directly produce two dimensional arrays of high K_u fct FePt nanoparticles for data storage. To conclude, pre-annealing pulsed laser exposure can be used to minimize the undesirable annealing effects of grain growth by lowering down the phase transition temperature of FePt thin films.

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