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Controlled grain growth in granular FePt–SiO_2 thin films under single pulsed laser anneals

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ABSTRACT

The high magnetocrystalline phase of $L1_0$ FePt has received considerable attention for achieving stable magnetization states in small volumes which could increase magnetic areal storage densities. When FePt is sputter-deposited, it adopts a magnetically soft A1 phase requiring annealing to phase transform to the $L1_0$ phase; this annealing results in detrimental grain growth which reduces the capacity for high areal storage densities. In the current work, a series of 10 nm thick granular FePt–SiO_2 thin films with various silica contents has been annealed at different fluences using a 10 ms pulse width, 1064 nm wavelength laser to determine if the silica matrix could inhibit this grain growth. The A1 to $L1_0$ phase transformation was confirmed by selected area electron diffraction. In general, the films annealed with approximately 25 J/cm^2 exhibited the highest $L1_0$ c/a tetragonality, 0.97, and coercivity of approximately 875 kA/m (11 kOe). For these films, the 38 vol.% silica incorporation resulted in a FePt grain size of approximately 8.5 nm as compared to 30 nm for films with no silica. The granular encasement of the FePt grains was effective at reducing but unable at inhibiting grain growth using single pulsed laser anneals.

1. Introduction

Increasing the storage capacity of magnetic media by increasing magnetic areal density necessitates a corresponding decrease in grain size. Since magnetization is proportional to the grain size, smaller grain sizes (*i.e. volumes*) can cause the magnetization to randomly fluctuate because of thermal energy effects, a phenomenon referred to as superparamagnetism. The superparamagnetic limit is a significant obstacle for achieving small grain, ultra-high areal storage density. The stability of small volumes or grains can be increased if materials with higher magnetocrystalline anisotropy, K_u are used. FePt with a $L1_0$ phase has received considerable interest for these applications because of its high K_u, which is $\approx 5\times10^6$ J/m^3 [1,2].

When FePt is sputter-deposited as a thin film, it adopts a metastable, solid-solution, face centered cubic structure (A1 phase) requiring subsequent anneals $>\approx$500 °C to phase transform into the thermodynamically stable and magnetically hard $L1_0$ phase [3,4]. During annealing, grain coarsening results and broadens the narrow grain size distribution required for ultra-high density storage applications [5,6]. Recently, we have reported that laser annealing of continuous FePt thin films using a variety of millisecond laser pulse widths can effectively phase transform FePt from A1 to $L1_0$ [7,8]. This study provided experimental confirmation that the phase transformation commences in the millisecond time regime [9,10]. However, even at the shortest pulse width of 2.5 ms, the ordered films experienced some amount of grain growth [7,8], although less than furnace thermal annealing for a given order parameter.

In this paper, the influence of co-sputtering the metallic alloy (FePt) with silica (SiO_2) was explored. The incorporation of an oxide has been commonly done to inhibit exchange coupling between magnetic grains [11]. The oxide is typically found in the grain boundaries and the encasement of each grain could provide a diffusion barrier for grain growth during the laser pulse anneal. This would then preserve the small grain sizes needed for ultra-high magnetic storage applications. Reduction of grain size and growth by oxide encasement has been done in conventionally annealed films [12], but these oxides tend to break-down during elevated temperature anneals resulting in the loss of the desired microstructure [13]. The outstanding question is whether laser annealing, with its millisecond annealing time scales, results in less grain growth and the preservation of the desired granular microstructure.

2. Experimental conditions

A series of 10 nm thick FePt–SiO_2 thin films was deposited onto 2-inch Si substrates by magnetron co-sputtering from Fe, Pt, and SiO_2 targets in an AJA ATC-1500 sputtering system. Prior to deposition, the chamber was evacuated by mechanical and turbo-mechanical pumps to $<10^{-5}$ Pa, at which point 10 standard cubic centimeters per minute

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of ultra high purity Ar was flowed into the chamber to 0.27 Pa (2 mTorr) to serve as the sputtering gas. The deposited film structure was Si_3N_4(5 nm)/FePt–SiO_2(10 nm)/Si_3N_4(5 nm)/Si substrate. A Si_3N_4 composite target was used to grow the Si_3N_4 barrier layers which prevented undesired thin film reactions with either the substrate or atmosphere during the anneal. Scanning transmission electron microscopy-X-ray energy dispersive spectroscopy (STEM–XEDS) analysis confirmed that the FePt composition was equi-atomic. The XEDS standard for the Cliff–Lorimer sensitivity k-factor was determined using a homogeneous FePt film whose composition was quantified by Rutherford backscattering. Post-deposition, the substrate with film was diced into 5 mm × 5 mm squares for the laser annealing studies.

An Electrox Scorpion Nd-YAG laser with a wavelength of 1064 nm was used to anneal the thin film specimens. The specimens were placed in a sample holder which had a hole through the center. The hole minimized the contact to <0.5 mm at the edges of the specimen to reduce thermal contact with the mount. The specimens were annealed in air using a single pulse with a pulse width of 10 ms. The pulse width was controlled by a mechanical shutter in the laser. The delivered laser fluence was varied from 0 to 45 J/cm^2. The specimens were placed 40 mm from focus such that the defocused laser energy spread had a Gaussian distribution with 20% varia-tion from the peak intensity in the center to the edge over 5 mm at this position [7]. To minimize potential delamination, *i.e.* thermal shock because of thermal expansion differences between the film and the substrate during the laser pulse, the specimens were pre-heated to a temperature of 200 °C using a quartz lamp positioned under the ceramic mount. This pre-heat temperature is below the A1 to $L1_0$ ordering temperature [10].

Transmission electron microscopy (TEM) was performed using a 200 keV Field Emission FEI Tecnai F20 Supertwin (S)TEM. The samples were prepared by standard backside Si polishing to <100 μm, dimpling to 15 μm and ion milling to electron transparency in a Gatan PIPS®. The grain size and grain size distribution, determined from approximately 300 manually traced grains in the bright field image for each analysis [14], was quantified using the NIKON NIS elements basic research software platform.

In-plane hysteresis loops were measured with a Princeton Instruments Model 2900 Alternating Gradient Magnetometer (AGM) with a maximum applied field of 1432 kA/m (18 kOe).

3. Results and discussion

The saturation magnetization, M_s, and average grain size, <D>, dependence of as-deposited FePt–SiO_2 films versus SiO_2 sputtering power is plotted in Fig. 1. The M_s values are defined with the applied field of 1432 kA/m. All FePt films were fully saturated with this applied field since they were in the magnetically soft, A1 phase state. As the SiO_2 sputtering power was increased, the M_s and <D> decreased. Plan view TEM images taken from the selected samples indicate that <D> was approximately 17.3 nm (for no silica addition) and 8.3 nm and 3.5 nm with increasing SiO_2 content. The TEM images clearly indicate a transition from a continuous film to a granular film with the silica addition. The volume fraction of silica was calculated from the M_s reduction using the no silica film as the reference value. The silica volume fraction was found to be 12 vol.% and 38 vol.% respectively.

The in-plane coercivity, H_c, dependence on the delivered energy fluence for 0, 12, and 38 vol.% SiO_2 content films is plotted in Fig. 2(a). As the delivered laser energy fluence increased, H_c increased to a maximum at ≈25 J/cm^2. Further increases in the energy fluence resulted in a decrease in coercivity. The increase in H_c corresponded to an increase in the tetragonality or the c/a lattice parameter ratio, plotted in Fig. 2(b), as determined by selected area electron diffraction (SAED). The maximum H_c corresponded to a c/a ratio near 0.97, which is nearly equivalent to a bulk fully ordered equi-atomic $L1_0$ FePt alloy [15].

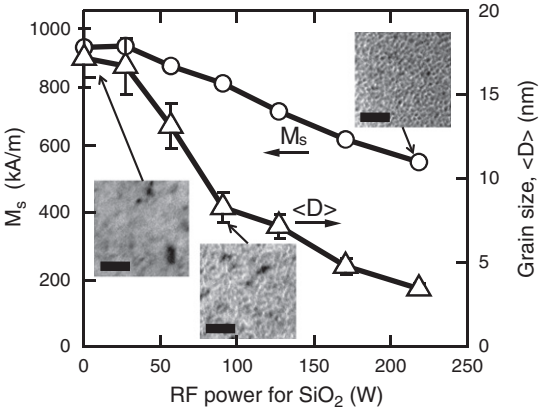

Fig. 1. Saturation magnetization, M_s, and grain size of as-deposited FePt–SiO_2 film with the A1 phase as functions of SiO_2 sputtering power. Fe and Pt power was kept constant during deposition. M_s values are defined with the magnetization with an applied field of 1432 kA/m (18 kOe). Films are fully saturated with this applied field since they have Al soft magnetic phase. Plan-view TEM micrographs taken from specific samples are also shown. The scale bar in TEM image represents 20 nm. The open circle is the M_s value and the open triangles are the average grain size.

As the delivered energy fluence increased, each film experienced grain growth, Fig. 3(a), but retained the granular microstructure for fluences less than ≈25 J/cm^2, Fig. 3(b). The extent of grain growth was less for films with increasing silica content. The inability to inhibit grain

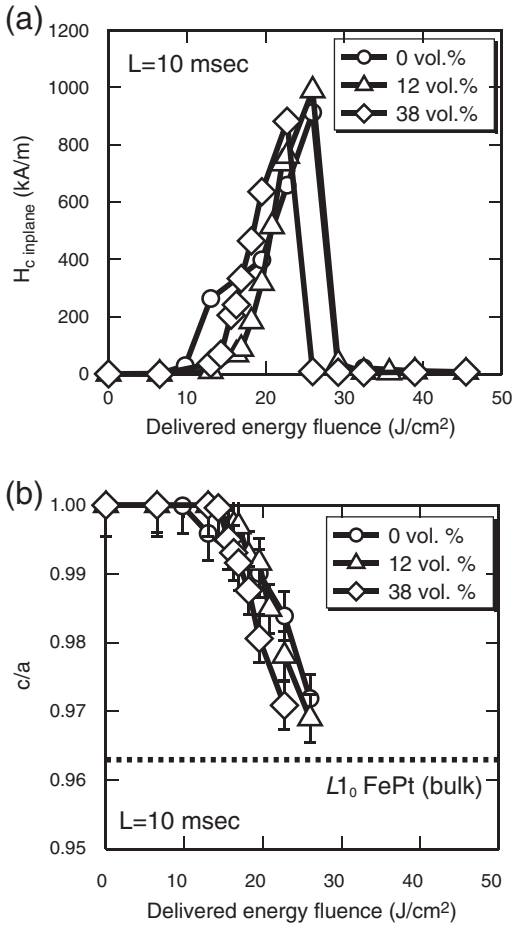

Fig. 2. (a) In-plane coercivity dependence on delivered energy fluence for various SiO_2 contents. Here, energy of '0' corresponded to the as-deposited state. (b) c/a ratio on delivered energy fluence for various SiO_2 content. c/a ratio of bulk $L1_0$ FePt phase [15] is also shown as the horizontal dashed line.