Thin Solid Films 517 (2009) 2753–2757

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Lowering of $L1_0$ phase transition temperature of FePt thin films by single shot H^+ ion exposure using plasma focus device

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ARTICLE INFO

Article history:
Received 10 April 2008
Received in revised form 4 November 2008
Accepted 13 November 2008
Available online 24 November 2008

Keywords:
FePt
Phase transition
Pulsed ion irradiation
Plasma focus
Magnetic properties
X-ray diffraction

ABSTRACT

FePt thin films are exposed to pulsed energetic H^+ ion beam from plasma focus. In irradiated films, the phase transition from the low K_u disordered face-centered-cubic structure to high K_u ordered face-centered-tetragonal phase was achieved at 400 °C with the order parameter S ranging from 0.73 to 0.83, high coercivity of about 5356 kA/m, high negative nucleation field of about 7700 kA/m and high squareness ratio ranging from 0.73 to 0.79. The advantage of using plasma focus device is that it can lower phase transition temperature and significantly enhance the magnetic properties by a pulsed single shot exposure.

1. Introduction

Ultra-high density magnetic recording media require a narrow size particle distribution and particle isolation to maintain the thermal stability and to reduce the media noise. However, if the size is reduced below critical size, particles become superparamagnetic. One way to lower the critical size is to use materials with higher magnetocrystalline anisotropy [1]. The $L1_0$ ordered equiatomic FePt alloy with large magnetocrystalline anisotropy constant K_u ($K_u = 6.6\times10^6$–1×10^7 J/m^3 and $M_s = 1140$ kA/m) [2] has been considered as one of the most promising candidates for high density magnetic recording media. However, the high annealing temperature needed for the phase transition from disordered face-centered-cubic (fcc) phase to chemically ordered face-centered-tetragonal (fct) structure $L1_0$ phase, which was reported to be greater than 500 °C [3,4], may cause agglomeration or grain growth and hence the particles no longer stay isolated [3].

There have been several attempts to lower the ordering temperature. The addition of a third element (Cu, Ag, and Au) into these alloys was reported to be effective for reducing the ordering temperature [5,6]. It was also reported that in multilayered Fe/Pt, Co/Pt, etc., their activation energy for ordering can be decreased [7,8]. Lin et al. [9] reported the lowering of the phase transition temperature to 300 °C in FePt:Al_2O_3 nanocomposite thin films synthesized by magnetic trapping assisted pulsed laser deposition. Another approach for lowering down the transition temperature is using ion irradiation. Ion irradiation has been applied widely not only to modify the magnetic properties in magnetic thin films or superlattice but also to lower the phase transition temperature [10–13]. He^+ irradiation was reported to control the degree of chemical ordering in FePt/FePd films [10,14]. The effects of ion (B^+, Cr^+, Ga^+, and Nb^+) irradiation on the crystalline structure and magnetic properties of $L1_0$ phase FePt films were investigated by Hasegawa et al. [11]. High energy (350 keV) He^+ irradiation was reported by Wiedwald et al. to lower the ordering temperature by more than 100 °C [12]. Moreover, the ordered $L1_0$ FePt phase was directly achieved by using continuous 2 MeV He^+ irradiation for about 1 h [13].

In this paper, we present the use of dense plasma focus as pulsed plasma driver for energetic H^+ ions irradiation of pulsed laser deposited FePt thin films to reduce the ordering transition temperature. The use of pulsed energetic ions of few hundreds of nanoseconds from plasma focus device, as opposed to that of long exposure time irradiation from continuous ion sources, provides a fast and effective way to lower down phase transition temperature with significantly enhanced magnetic properties.

2. Experiments

FePt thin films were grown on Si (001) substrates at room temperature by pulsed laser deposition in vacuum (better than 3×10^{-3} Pa). Continuum Nd:YAG laser (532 nm, 10 Hz, 10 ns and 80 mJ) was focused on FePt (50:50 at.%; Kurt J. Lesker, 99.99%) target disc with energy density of about 1.02×10^3 J/cm^2. The highly energetic

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0040-6090/$ – see front matter © 2008 Published by Elsevier B.V.
doi:10.1016/j.tsf.2008.11.113

H^+ ions, used in this investigation for the irradiation of FePt thin films, are obtained using a pulsed plasma driver called the dense plasma focus (DPF). We used a single-capacitor (30 μF) based UNU/ICTP (United Nation University/International Center for Theoretical Physics) plasma focus device charged to 14 kV, with storage energy of 2.94 kJ, resulting in the formation of short-lived hot and dense (10^{25}–10^{26} m^{-3}) plasma [15]. The working gas used is hydrogen, and the gas pressure was kept at 5×10^2 Pa. The various sub-systems of the device are shown in Fig. 1. The as deposited FePt thin film samples, as shown in Fig. 1, were affixed to the substrate holder placed along the anode axis at different distances from the top of the copper anode. An aperture assembly is employed, between the anode top and the thin film sample, to stop the plasma shock wave and to eliminate the possibility of contamination of the films by copper impurity coming from electron bombardment of copper anode rim. The FePt thin film samples were exposed to H^+ ion at different distances of about 5, 6 and 7 cm to investigate the effects of different ion flux on the magnetic properties and phase transition temperature. After irradiating as deposited thin films to single plasma focus shot, the samples were heated to the annealing temperature of 400 °C at the rate of 60 °C/min in vacuum and maintained at the annealing temperature for 1 h then cooled down naturally. The annealing temperature was finalized to 400 °C after making initial studies on samples irradiated at 5 cm where we found that it was minimum temperature at which fct phase in FePt could be induced. No phase transition was observed at temperatures below 400 °C and since we wanted to investigate the effects of irradiation parameters by changing the distance of irradiation on the phase transition properties of FePt films we kept the annealing temperature fixed at 400 °C. X-ray diffraction (XRD) patterns were measured by a Rigaku D/MAX-rA X-ray diffractometer with CuKα radiation. A Lakeshore 7400 vibrating sample magnetometer with maximum applied field of $10{,}000\times(4\pi)^{-1}$ kA/m was used to record in-plane hysteresis loops at room temperature to investigate the magnetic properties of the samples.

3. Results and discussion

Fig. 2 shows XRD patterns of the as-deposited and irradiated FePt thin film samples before and after annealing using θ–2θ scans. All samples exhibited preferred orientation along (111) diffraction plane. The as-deposited thin film, refer Fig. 2(a), exhibits fcc phase with very small crystallite size and weak crystallinity indicated by broad and weak peak of (111) plane at about $2\theta=40.8°$. The crystallite size calculated from Scherrer's formula [9], $t=0.9\lambda/(B\cos\theta_B)$, was found to be 3.3 nm. After 1 h annealing at 400 °C, the sample still remains in fcc phase with diffraction spectrum showing (111) and (200) fundamental peaks. The thermal annealing improved the crystallinity of the sample as noticed by the significant increase in (111) peak intensity. The crystallite size was found to increase to 9.6 nm, an expected effect of annealing.

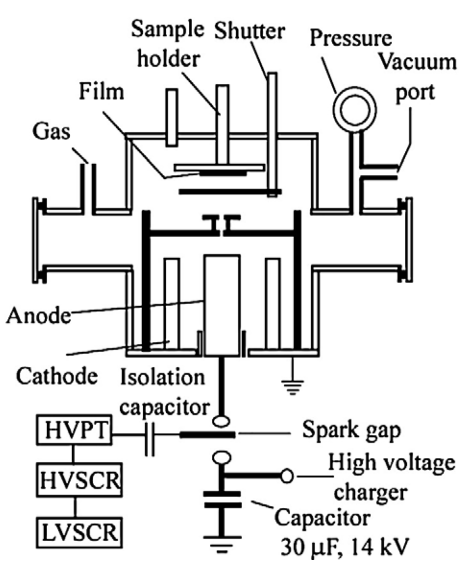

Fig. 1. Schematic of plasma focus device.

The XRD patterns of the samples irradiated at 5, 6 and 7 cm, without being annealed, are shown in Fig. 2(c, e and g). It may be noted that these samples still remain in fcc phase though the degree of crystallinity has been improved. The increase of crystallinity can be attributed to the processing of FePt thin film by the energetic H^+ ions. Sanchez and Feugeas [16] reported that energetic ions of plasma focus can transfer enough energy at a fast rate to the irradiated sample surface which rapidly achieves high temperature resulting in rapid local melting and quenching. Hence, we can say that pulsed ion beam in plasma focus provides transient heating/cooling of the exposed samples resulting in improvement in crystallinity of irradiated sample. Since the pulsed ion beam causes transient heating and rapid quenching of the sample surface, the average temperature rise of the exposed sample is negligible. The average temperature rise of sample surface by energetic pulsed ion exposure in plasma focus device was estimated by Rawat et al. [17]. They reported that the average surface temperature (as measured by k-type thermocouple interfaced to computer) of the sample surface after 30 exposure shots increased from 23.6 to 25.7 °C and hence one can easily conclude that the average temperature rise of the sample for one shot exposure is negligible. Since the ion and energy flux (and hence the corresponding ion irradiation heating) in plasma focus decrease with the increase in the irradiation distance[15], it explains the reduction in the degree of crystallinity (indicated by reduction in (111) peak intensity) for the samples irradiated at higher distances. The crystallite sizes for the samples irradiated at 5, 6 and 7 cm are calculated to be 7.2, 7.0 and 6.9 nm. The increase in crystallite size of irradiated sample as compared to that of as-deposited sample confirms plasma focus based pulsed ion irradiation transient heating. The decrease in crystallite size with increasing distance of irradiation can be attributed to lower ion irradiation heating at higher distance due to reduced ion and energy flux. Another noticeable feature in diffraction patterns of irradiated samples is the shift of (111) fundamental line to lower angle side to about $2\theta=40.6°$ as compared to that of $2\theta=40.8°$ for as-deposited sample. This indicates the increase in d-spacing of (111) planes from 0.220 nm for as-deposited sample to about 0.222 nm for ion irradiated samples which is due to the implantation of H^+ ions into the interstitial sites of fcc lattice of FePt causing the lattice to expand.

The XRD patterns of the irradiated samples after being annealed at 400 °C for 1 h are shown in Fig. 2(d, f and h). The $L1_0$ ordering phase has been achieved on all these samples which is indicated by the appearance of the (001)/(110) superlattice peaks and by the peak splitting (200)/(002) of fundamental peaks due to the tetragonal lattice distortion [5]. The lower annealing temperature for achieving $L1_0$ ordered fct phase for ion irradiated sample has been explained by Wiedwald et al [12]. According to them, one of the ways to lower down the annealing temperature at which the structural phase transition to $L1_0$ phase occurs is to reduce the activation energy, E_D, for diffusion say by increasing the number of defects in the crystal structure. The energetic ion irradiation can cause both the vacancy (by knocking the parent atom from the lattice site) and interstitial (by implantation of irradiating ions) point defects to reduce the activation energy of diffusion.

The annealing of irradiated samples was found to increase the crystallite size further to 16.1, 15.6 and 15.3 nm from 7.2, 7.0 and 6.9 nm for samples irradiated at 5, 6 and 7 cm respectively. It may also be noted that upon annealing the position of (111) peak for irradiated samples shifted to higher angles indicating the reduction in d-spacing. This reduction in d-spacing of ion irradiated samples upon annealing