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Diffusion of Au and its influence on the coercivity of $[FePt/Au/FePt]_{2x}$ thin films during annealing in different atmospheres

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ABSTRACT

Influence of heat treatment atmosphere on magnetic properties of post-annealed [FePt(15 nm)/Au(7.5 nm)/FePt (15 nm)]$_{2x}$ thin films was investigated. Thin films were deposited by dc magnetron sputtering at room temperature onto SiO_2(100 nm)/Si(100) substrates and post-annealed in a temperature range of 500 °C–800 °C for 30 s in flowing Ar and Ar + H_2 (3 vol%) atmospheres. It was shown that hydrogen addition into annealing atmosphere allows to achieve high value of coercivity ~1671.1 kA/m after annealing at 600 °C. Such coercivity could be reached by heat treatment in pure Ar just after annealing at 700 °C. This effect could be related to acceleration of Au grain boundary diffusion into $L1_0$-FePt phase. Such acceleration could be possible due to two reasons: hydrogen incorporation into Au crystal lattice, leading to interatomic bounds break and point defects formation; and hydrogen segregation at most defective areas, leading to cohesive energy decrease and upraise of significant stresses. As a result, nonmagnetic grid in grain boundaries of ferromagnetic material was formed at temperature lower by 100 °C.

1. Introduction

FePt based thin films are attractive materials for ultrahigh density magnetic recording application due to their excellent magnetic properties [1–4]. However, for high coercivity provision it is necessary to apply high temperature heat treatment in order to form ordered $L1_0$-FePt phase from initially disordered $A1$-FePt phase [5]. Grain growth occurs during heat treatment, leading to decrease of potential recording density. There are several approaches to increase of $L1_0$-FePt phase ordering degree and consequently its coercivity at lower annealing temperatures.

Variation of FePt films annealing atmosphere composition could lead to control of phase transformations kinetics and temperature intervals, structural properties, surface roughness and thereafter affects the magnetic properties. For instance, H.H. Hsiao et al. showed that nitridation of $Fe_{53}Pt_{47}$ films at 300 °C leads to lattice expansion of ordered phase and increase of films coercivity [6]. Particularly, it was determined that presence of H_2 in inert annealing atmosphere leads to thermal stabilization of FePt films' grains size and surface roughness [7].

Introduction of nonmagnetic layers into FePt based stacks is another well-known approach. In this case ordering process could be enhanced because of stresses, arising from differences in layers' lattice constants and thermal expansion coefficients. As nonmagnetic layers, Ag [8,9], AgCu [10], Cr and Cu [11], C [12], etc. could be used.

Application of Au in stacks with layers of FePt alloy [13] or Pt/Fe separate metals [14] was also reported to be effective. It was shown that in order to prevent the magnetic coupling, arising from the interconnected $L1_0$-FePt grains and increase films coercivity, it is required to surround ferromagnetic FePt grains by nonmagnetic component [15,16]. For instance, F.T. Yuan et al. [17] showed that drastic increase of $L1_0$-FePt phase coercivity from 1193.7 kA/m to 1591.6 kA/m could be reached by annealing of Au/FePt bilayered stacks at 800 °C for 1 h. Au diffusion leads to $L1_0$-FePt phase grains separation, resulting in magnetic grains decoupling. Positive impact of Au addition on properties of FePt nanoparticles was also shown in [18]. Increasing of Au concentration leads to increase of tetragonality, chemical ordering degree and as a consequence to rise of FePt nanoparticles coercivity. Increase of coercivity of $[FePt/Au]_{20}$ stacks, heat treated at 300 °C, with increase of Au layer thickness was also reported in [19].

In this work we propose combined approach for control of structural properties and coercivity of FePt thin films by means of introduction of additional Au layers and variation of heat treatment atmosphere (annealing in Ar and forming gas (Ar + H_2)). We assumed that in this case

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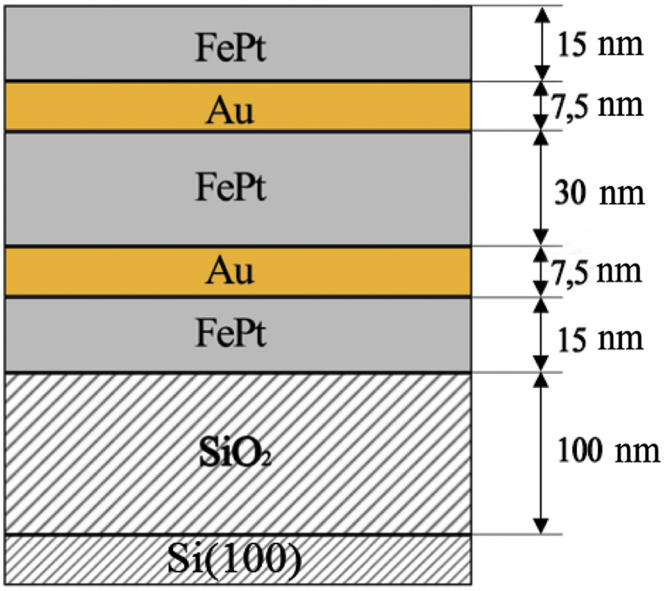

Fig. 1. Schematic illustration of $[FePt(15\,nm)/Au(7.5\,nm)/FePt(15\,nm)]_{2x}$ thin film layered structure.

it could be possible to reach optimal ratio between ordering degree and Au atoms grain boundary diffusion velocity with blocking of re-crystalization processes.

2. Experimental

$[FePt(15\,nm)/Au(7.5\,nm)/FePt(15\,nm)]_{2x}$ thin films were deposited by dc magnetron sputtering onto thermally oxidized Si(100) substrates at room temperature. FePt layers were obtained by co-deposition from Fe(99.9%) and Pt(99.95%) targets, and Au layers were deposited using Au(99.9%) individual target. Power of 20 W, 9 W and 15 W was applied to Fe, Pt and Au targets, respectively. Distance between targets and substrate was adjusted to 17 cm. The Ar sputter pressure was adjusted to 0.35 Pa for all depositions. Base pressure of deposition chamber was $5 \times 10–^5$ Pa. Calculated growth rates were 0.03 nm/s and 0.05 nm/s for FePt and Au layers, respectively. Thickness of the layers was verified by Rutherford backscattering spectroscopy.

Fig. 1 shows the schematic illustration of layered structure of the investigated thin films. Relatively high thickness of Au layers was chosen in order to provide infinite diffusion source for uniform saturation of FePt phase grain boundaries. $[FePt/Au/FePt]_{2x}$ films with double repetition of the layers were prepared in order to increase quantity of interfaces which are origins of additional mechanical stresses. Such stresses could lead to diffusion and ordering enhancement [17].

Post-annealing of the film samples in temperature range of 500 °C–800 °C was carried out in Ar and Ar + H_2 (3 vol%) atmospheres with a flowing speed of 0.2 *l*/min and annealing duration of 30 s, using a fixed heating rate of 10 °C/s. Heat treatment temperature interval of 500 °C–800 °C was chosen with reference to [7].

Phase composition and structural properties of the as deposited and post-annealed films were investigated in the SPring-8 synchrotron radiation facility, using the RIKEN Materials Science beamline BL44B2 [20] by the grazing-incidence wide-angle X-ray scattering (GIWAXS) method. Measurements were carried out in geometry of 0.5° incident angle to the sample surface. In GIWAXS measurements, the X-ray of wavelength 1.08 Å from BL44B2 was used. Vertical and horizontal beam sizes were adjusted to 0.01 mm and 3.0 mm, respectively. The sample was set to the Debye-Scherrer camera equipped with an off-line imaging plate ($400 \times 200\,mm^2$), with ranges from 2° to 78° at 2θ intervals of 0.01°.

Investigation of annealed thin films surface morphology was done by atomic-force microscopy (AFM). Magnetic properties were measured by superconductive quantum interference device-vibrating sample magnetometry (SQUID-VSM). Magnetization curves were measured at room temperature in two geometries: applying magnetic field up to 5570.4 kA/m in the film plane and out of the film plane.

Chemical depth profiling was done by secondary ions mass-spectrometry. Argon ions with energy of 5 keV were applied as a primary beam, current of ion beam discharge was 0.4 mA and current of the primary beam was 15 μA (calculated current density – 2.5 $\mu A/mm^2$). Analysis of results was done using Spectrum Recorder software. Ta masks with a thickness of 0.1 mm and hole diameter < 1 mm were used in order to increase depth resolution. Focusing of the primary Ar^+ ion beam was performed by means of optically aligned laser pointer.

3. Results and discussion

3.1. Magnetic properties

Magnetic *M-H* hysteresis loops of $[FePt(15\,nm)/Au(7.5\,nm)/FePt(15\,nm)]_{2x}$ films after annealing in Ar and Ar + H_2 atmospheres at different temperatures are shown in Fig. 2. It is worth to mention that there is almost constant remanence ratio at chosen temperature interval after annealing in both investigated atmospheres. After annealing at 500 °C in Ar + H_2 atmosphere films' coercivity reaches 1193.7 kA/m, after annealing in Ar atmosphere at the same temperature – 875.4 kA/m (Fig. 2, a). Increasing of annealing temperature up to 600 °C leads to more pronounced difference between coercivity values (Fig. 2, b). After annealing at 600 °C in Ar atmosphere coercivity reaches value of 931.1 kA/m, after annealing in Ar + H_2 mixture – 1671.1 kA/m. Increase of coercivity of FePt films without additional layers during rising of annealing temperature is related to increase of $L1_0$-FePt phase quantity and ordering degree. So it can be assumed that higher value of coercivity after annealing in Ar + H_2 mixture could be related to positive hydrogen impact on $L1_0$-FePt phase chemical ordering. At the same time, reached coercivity value of 1671.1 kA/m evidences the influence of not only hydrogen but Au additional layers as well. It should be noted that annealing in Ar atmosphere at 600 °C of epitaxial $[FePt(1\,nm)/Au(0–2\,nm)]_{10}$ stacks leads to coercivity of 1193.7 kA/m [13].

Annealing in Ar atmosphere at 700 °C leads to drastic increase of coercivity up to 1671.1 kA/m, after annealing in forming gas at the same temperature coercivity is 1830.8 kA/m. So it can be concluded that the most pronounced difference in coercivity values after heat treatment in investigated atmospheres is determined after annealing at 600 °C. Also, there is some shoulder on the corresponding *M-H* loop that could indicate possible existence of soft-magnetic fcc FePt phase. However, there are no peaks from *A1*-FePt phase on corresponding diffraction patterns (Fig. 3). Probably it could be caused by very small amount of disordered phase in the film after annealing at 500 °C. Such a small amount could be enough for influence on the magnetic behavior, but not enough to be registered by XRD.

Coercivity value depends not only on chemical ordering degree but on grains size and amount of nonmagnetic component in grain boundaries, which is controlled by diffusion velocity affected by annealing atmosphere [21]. Let's overview main parameters affected coercivity during annealing of investigated layer stacks in different atmospheres.

3.2. Structural properties

X-ray diffraction patterns obtained using synchrotron radiation are shown in Figs. 3, 4. Superstructure (001) peak from chemically ordered $L1_0$-FePt phase can be clearly seen on all diffraction patterns from the films after annealing in both investigated atmospheres.

Second order of (001) $L1_0$-FePt phase superstructure peak – (002) – is present on all diffraction patterns as well. It could be concluded that