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Physics Procedia 54 (2014) 23 – 29

Physics Procedia

International Conference on Magnetic Materials and Applications, MagMA 2013

Magnetic properties of FePt based nanocomposite thin films grown on low cost substrates

A. Gayen[a], B. Biswas[a], A.K. Singh[a], P. Saravanan[b], and A. Perumal[a,]*

[a]*Department of Physics, Indian Institute of Technology Guwahati, Guwahati 781039, India*
[b]*Defence Metallurgical Research laboratory, Hyderabad 500058, India*

Abstract

We report a systematic investigation of temperature dependent magnetic properties of FePt single and FePt(30)/M/Fe(5) nanocomposite thin films prepared by sputtering technique on low cost substrates at ambient temperature and post annealed at different temperatures. With increasing annealing temperature, $L1_0$ ordering, hard magnetic properties and thermal stability of FePt films are improved. The formation of interlayer exchange coupling between hard and soft magnetic layers in FePt/M(Al,Cu,C)/Fe films depends strongly on interlayer materials and interface morphology. A strong interlayer exchange coupling was achieved when the C interlayer thickness was about 0.5 nm, which enhances saturation magnetization largely. Also, the magnetization reversal process changes from incoherent to coherent switching process, which results a single hysteresis loop. High temperature magnetic studies revealed that the effective reduction in the coercivity decreases from 34 Oe/K to 13 Oe/K by the introduction of a thin C(0.5 nm) layer in FePt/C/Fe film. This reveals a promising approach to improve the stability of hard magnetic properties at high temperatures, suitable for high temperature magnetic applications.

Peer-review under responsibility of Department of Physics, Indian Institute of Technology Guwahati

keywords— Nanocomposite; Interlayer exchange coupling; Multilayer film; Coercivity; Magnetization reversal process.

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1. Introduction

Futuristic magnetic devices such as biasing nanomagnets and exchange coupled nanocomposite magnet in micro

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* A. Perumal. Tel.: +91-361-2582714; fax: +91-3612582749/ 2690762.
E-mail address: perumal@iitg.ernet.in

Peer-review under responsibility of Department of Physics, Indian Institute of Technology Guwahati
doi:10.1016/j.phpro.2014.10.032

electromagnetic devices, and ultra-high-density magnetic storage media need enhanced performance of hard magnetic thin films [1]-[2]. Single domain magnetic particles with high magnetic anisotropy energy are expected to play a major role in improving the hard magnetic properties and to attain large thermal stability. Hence, extensive research work has been carried out recently to search for suitable thin films with magnetic alloys having large magnetic anisotropy energy from the standpoint of high density magnetic recording technology and permanent magnet applications [2]-[3]. Recently, $L1_0$ ordered equiatomic FePt based alloys are considered as one of the promising candidates for hard magnet applications because of its extremely high magnetic anisotropy energy ($K_u \sim 10^8$ erg/cc). Since a high K_u material can produce a large H_C, attempts were shown to fabricate FePt based alloys in various forms [4]-[6]. However, the maximum energy product is limited by saturation magnetization (M_S) and hence the permanent magnet with high-energy product is not achieved mainly because of a trade-off in obtaining the large coercivity (H_C) and high M_S. On the other hand, Coehoorn et al [7], and Kneller et al [8], proposed the exchange spring magnet as a way to create a next generation high performance permanent magnet and magnetic recording media. Since then many investigations have been carried out on various hard (Sm-Co, FePt and Co-Pt) and soft (Fe, Co, Fe-Co, Fe_3Pt) ferromagnetic (FM) phases by coupling the soft FM phase directly to the hard FM phase and studied the effects of bilayer and multilayer structures and various heat treatment process on the improvement of properties of nanocomposite magnets [9]-[12]. The maximum energy product of 52.8 MGOe was achieved experimentally by Liu et al [13], in FePt/Fe_3Pt nanocomposite thin film.

Recently, interlayer exchange coupling using two FM films, hard and soft magnets, separated by a thin nonmagnetic layer, resulting in an indirect exchange coupling between the FM layers was reported to construct an exchange spring [14]. The advantages of such coupling are that the coherent interface is not necessary and the interface diffusion may not have serious effects on the exchange coupling. In order to use as a desirable magnet and for the application of high density magnetic recording media, both intrinsic (high M_S, high Curie temperature, and high K_u) and extrinsic (high energy product, high H_C, thermal stability and corrosion stability) properties should be characterized. A careful review on the literature of FePt films reveals that the FePt films exhibits a large coercivity (> 30 kOe) in very thin island-like films and particular films [2],[15], but the coercivity drops drastically when they become continuous [6],[16]. These results reveal that the fundamental magnetic properties of FePt films are very sensitive to the chemical ordering and microstructure. Also the FePt films with high $L1_0$ ordering show very high H_C and the introduction of proper exchange coupling using soft magnets improves the high energy product. However, only a few reports have been published on the role of chemical ordering on thermal stability and high temperature magnetic properties of FePt based single domain particles [17], but no detailed investigations on indirect exchange spring have been reported. Therefore, we report (i) high temperature magnetic properties of FePt films fabricated on the low cost substrates with different chemical ordering, (ii) the formation of interlayer exchange coupling, and (iii) their effects on the stability of magnetic properties at high temperature by producing FePt(30 nm)/M(y nm)/Fe(5 nm) nanocomposite thin films with M – Al, Cu and C as a nonmagnetic layer.

2. Experimental Details

FePt (x nm) films with x = 10 – 50 nm were prepared by DC magnetron sputtering technique using a high purity Fe target with Pt pellets on it on low cost substrates (Thermally oxidized Si and Si(111) wafers). This provides a homogenous single layer Fe-Pt film and the composition was altered by adjusting the number of Pt pellets on Fe target. Subsequently, a series of films with FePt(30 nm)/M(y nm)/Fe(5 nm) (M = Al, Cu, and C, and y = 0, 0.25, 0.5, 1, 2 nm) were deposited by sputtering FePt, M, and Fe targets at ambient temperature. As-deposited films were post deposited at different temperatures (450 and 550 ^oC) for 45 minutes under a high vacuum. This post annealing process orders the face centered cubic of FePt into face centered tetragonal structure. The base pressure of the chamber was better than 3×10^{-5} Pa and the high purity argon of 10 mTorr for FePt and Fe, 20 mTorr for C and 8 mTorr for Cu and Al was flown during the sputtering. The nominal thicknesses of the FePt, M, and Fe films were controlled based on the pre-calibrated sputtering rates of the FePt, M and Fe, respectively. Crystal structure were examined by X-ray diffractometer (Rigaku TTRAX 18 kW) with Cu-K_α radiation (λ = 1.5405 Å). Composition of the FePt films was examined by energy dispersive X-ray spectroscopy attached to a scanning electron microscope (SEM, Leo 1430VP). Temperature dependent magnetic properties of the films were analyzed by using Vibrating Sample Magnetometer (VSM, Model: LakeShore 7410, USA).

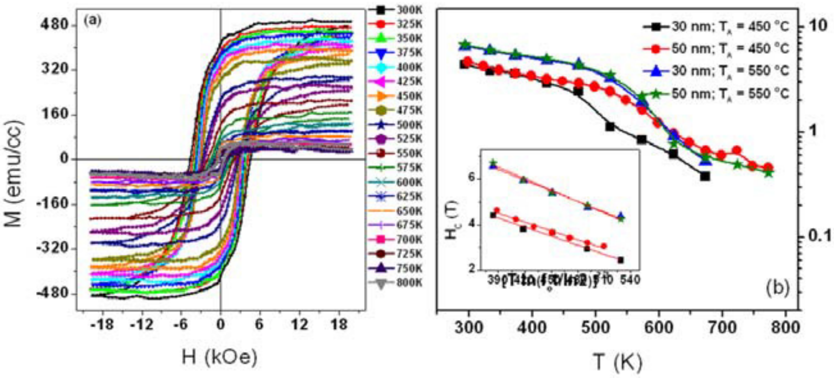

Fig. 1. (a) Temperature dependent $M - H$ loops of FePt(50 nm) film annealed at 450 °C; (b) Variations of coercivity with temperature for FePt films with 30 and 50 nm thicknesses. Inset: Coercivity as a function of [T ln(f_0t/ln2)]$^{2/3}$ for the FePt films annealed at different temperatures.

3. Results And Discussion

Fig.1 displays a typical $(M - H)_T$ loops obtained at different temperatures for the FePt(50 nm) film annealed at 450 °C and the variations of temperature dependent coercivity ($H_C(T)$) for the FePt films with 30 and 50 nm thicknesses. While the shape of the $M - H$ loops measured at different temperatures are similar, the saturation magnetization (M_S) and H_C decrease with increasing temperature. This is mainly due to the reduction in the magnetocrystalline anisotropy energy with increasing temperature. On further increasing the temperature above 600 K, $H_C(T)$ decreases largely due to instability in the $L1_0$ ordered state and finally they exhibit soft magnetic properties. The extracted values of H_C for the different FePt film thicknesses are plotted as function of temperature in Fig. 1(b). The room temperature H_C shows a strong dependence of annealing temperature, which could be attributed to the increase in the chemical ordering of $L1_0$ phase. The improvement in the chemical ordering was also confirmed from the structural analysis (not shown here). Also, $H_C(T)$ shows a substantial thickness dependent for the films annealed at low temperature, while the films annealed at high temperature did not show much difference and follow a similar trend for different film thicknesses. This can be attributed to the size dependent chemical ordering in the FePt films with different thicknesses [18]. To understand the effect of post annealing on the thermal stability, $H_C(T)$ data were analyzed using Sharrock's equation [19] given as

$$H_r(t,T) = H_o\left\{1 - \left[\left(\frac{k_B T}{E_b}\right)\ln\left(\frac{f_o t}{\ln 2}\right)\right]^n\right\} \tag{1}$$

where, H_r is the remanent coercivity dependent upon the field exposure time t and temperature T. H_o is the intrinsic coercivity without thermal agitation, k_B is the Boltzmann's constant, f_o is the attempt frequency, t is time of hysteresis measurement and E_b is the energy barrier at zero field. The value of n is taken as 2/3 due to slightly unaligned particles [20]. The attempt frequency f_o and the hysteresis measurement time are taken as 10^{10} Hz and 1 second, respectively. Inset of Fig. 1(b) displays the applicability of Sharrock's equation on $H_C(T)$ data for FePt films. The fitting resulted the energy barrier values of 1.40 *eV* and 1.61 *eV* for the FePt films of 30 and 50 nm annealed at 450 °C, and 1.96 *eV* and 2.02 *eV* for the FePt films of 30 and 50 nm annealed at 550 °C, respectively.

This reveals the corresponding thermal stability [the ratio between the energy barrier (E_b) to thermal energy (k_BT) at room temperature] of about 54 and 63 for the FePt films of 30 and 50 nm annealed at 450 °C and 76 and 78 for the FePt films of 30 and 50 nm annealed at 550 °C, respectively. These results confirm that the values of energy barrier increase considerably with the annealing temperature due to the improvement in the $L1_0$ chemical ordering. Since the FePt films annealed at 550 °C show enhanced properties, we have taken FePt(30 nm) film for the study of FePt based nanocomposites.

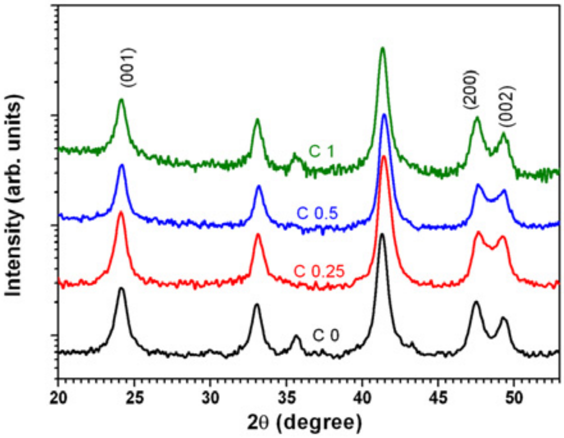

Fig. 2. XRD patterns for FePt(30 nm)/C (x nm)/Fe(5 nm) films with $x = 0 – 1$ nm prepared on Si(111) substrates at ambient temperature and post annealed at 550 °C.

To investigate the exchange spring in the FePt film and to study their temperature dependent magnetic properties and the thermal stability, a series of FePt(30 nm)/M(x nm)/Fe(5 nm) trilayer films with $x = 0$, 0.25, 0.5, and 1 nm and different intermediate layers using Cu, Al, and C were fabricated on Si(111) substrates. Fig. 2 displays the typical X-ray diffraction (XRD) patterns of trilayer films with different C thicknesses deposited at ambient temperature and post annealed at 550 °C. It is evident from the figure that superlattice diffraction line of (001) is observed around $2\theta = 24°$ and all the films are $L1_0$ ordered with *fct* structure after post annealing. The other unlabeled diffractions peaks are due to the Si substrate. It may be noted that no XRD patterns showed any diffraction peaks related to Fe or spacer layer. Fig. 3 shows the room temperature $M – H$ loops of FePt(30)/M(x)/Fe(5) films and the extracted values of H_C, M_S and the field necessary to saturate the films magnetization (H_{sat}). In the case of bi-layer films, $M – H$ loop exhibits a clear kink in second quadrant due to an incoherent or separate magnetic reversal process suggesting that there is a weak FM coupling or no coupling between the hard and soft layers [9]. The introduction of Cu or Al spacer reduces the kink observed in bilayer film gradually up to 1 nm without a much change in M_S values. In addition, the magnetic field required for saturating the films (H_{sat}) and H_C decrease significantly with increasing the Cu/Al layer thickness. Interestingly, the introduction of C interlayer exhibits quite different properties: (i) With increasing the C to 0.25 nm, the kink in the second quadrant slightly reduces, but H_{sat} decreases noticeably. (ii) When the C thickness is increased to 0.5 nm, the kink in the loop disappears and $M – H$ loop exhibits a single hysteresis with almost rectangular shape. This confirms a strong interlayer FM exchange coupling between the FePt and Fe layers through C interlayer. (iii) M_S value has increased to 990 emu/cc remarkably. (iv) On further increasing the C layer thickness, the shape of the $M – H$ loop and values of M_S remain same. The values of H_C and H_{sat} decrease gradually with increasing C interlayer thickness. To understand the effect of interlayer thickness on the magnetic parameters, the values of H_C, H_{sat}, and M_S were extracted from the loops and depicted in Fig.3(e) and (f), respectively for FePt(30)/M(x)/Fe(5) nanocomposite films. It is observed that H_C and H_{sat} decrease largely with increasing C thickness up to 1 nm. On the other hand, M_S values increases quickly to 990 emu/cc around 0.5 nm. For the Cu/Al interlayer, both H_C and H_{sat} exhibit a slow decrease and M_S remains almost contsnt with increasing the Al/Cu thicknesses. The observed results suggest that the

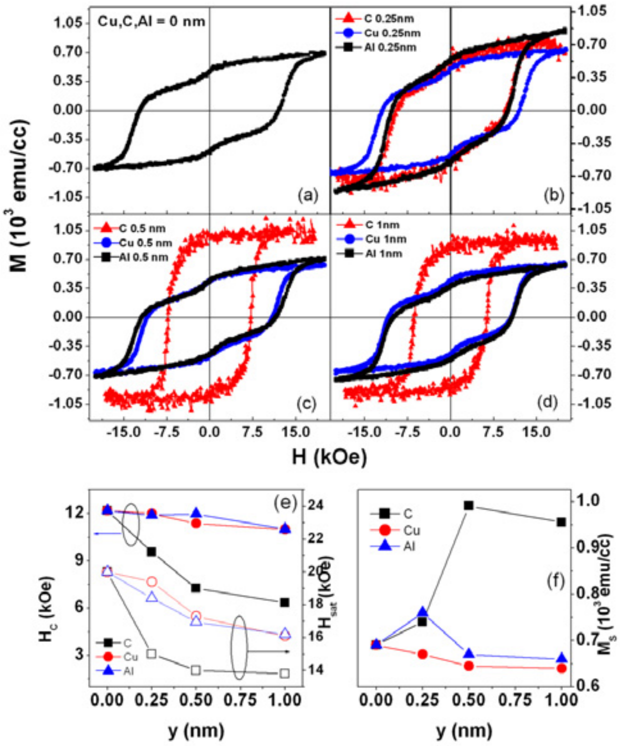

Fig. 3. (a-d) Room temperature $M-H$ loops, and (e) Variations of H_C and H_{sat} (e) and M_S (*f*) of of FePt(30 nm)/M (*x* nm)/Fe(5 nm) films with M – Al, Cu and C and $x = 0-1$ nm prepared on Si(111) substrates at ambient temperature and post annealed at 550 °C.

interlayer FM exchange coupling between the FePt and Fe layers strongly depends on the type of materials used as interlayers and their thickness, and interface morphology [14], [21]. All the trilayer films were prepared at room temperature and post annealed at 550 °C. It is known that the post annealing may cause a possible inter diffusion between the layers in multilayer films resulting a change in the interface morphology [22]. Also, the growth morphology of FePt films with annealing may provide inhomogeneous interface morphology to subsequent layers [21]. Since the trilayer films with Cu/Al interlayer did not show a considerable change in the magnetic properties, this may be correlated to the possible inter diffusion of Cu either into FePt or Fe resulting an unclear interface between hard and soft layers, [22]. Although the kink observed in the second quadrant weakens slowly, the formation of interlayer exchange coupling was not at all observed. On the other hand, the introduction of C between the hard and soft phases is expected to provide stable interfaces even after annealing [23]. Hence, the hard and soft phases are clearly separated by the C interlayer, which induces the interlayer exchange coupling resulting a coherent switching process and giving rise to a single hysteresis loop. Rheem et al [23], reported that the robust interlayer exchange coupling obtained in the FePt(30)/FeCo(10)/FePt(30) films is improved initially by introducing a thinner C/Ta interlayer, but eventually degraded at larger thicknesses due to the formation of continuous TaC films between FM layers. Jiang et al [14] reported the indirect exchange between FePt and Fe using Ru interlayer, as observed FePt/C/Fe trilayer films.

To understand the effect of interlayer exchange coupling on the stability of the hard magnetic properties, $(M-H)_T$ loops were observed at different constant temperatures and the extracted values of $H_C(T)$ are plotted in Fig. 4 for different Cu and C interlayers. To correlate the effect of interlayer exchange coupling on the magnetic properties, we have carefully analyzed the effective variation of $H_C(T)$ data for various interlayer thicknesses. It is revealed that the effective reduction in the H_C decreases from 34 Oe/K for $x = 0$ to 25 Oe/K for FePt/Cu(1)/Fe films. For the FePt/C(x)/Fe films, H_C reduces from 34 Oe/K for $x = 0$ to 13 Oe/K for $x = 0.5$ nm. This provides clear evidence that the interlayer FM exchange coupling between the hard and soft phases through a non-magnetic film plays a major control on the stability of the hard magnetic properties of FePt films at higher temperatures leading to a sluggish decrease in H_C. Furthermore, we need a careful analysis of $H_C(T)$ study at different Fe layer thicknesses in FePt/C/Fe trilayer films to reveal a detailed information on the effects of interlayer exchange coupling and their possible applications in permanent magnets.

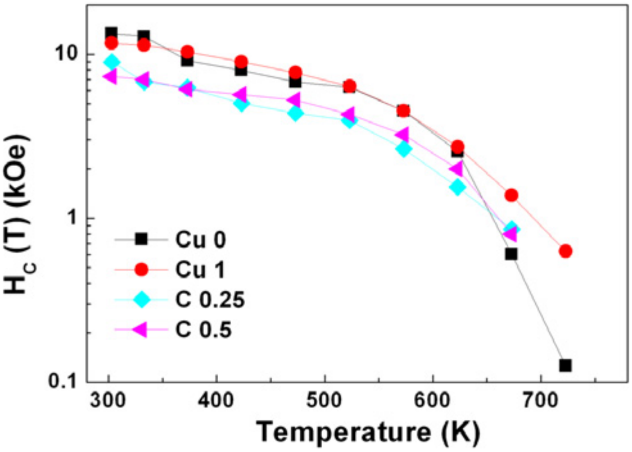

Fig. 4. Variations of coercivity with temperature for FePt(30 nm)/M (x nm)/Fe(5 nm) trilayer films with M – Cu (0 and 1) and C (0.25 and 0.5 nm) prepared on Si(111) substrates at ambient temperature and post annealed at 550 °C.

4. Conclusion

We have studied the high temperature magnetic properties of FePt single layer thin films and FePt/M/Fe nanocomposites through various interlayers at different interlayer thicknesses. It is perceived that the interlayer exchange coupling between the hard and soft magnetic phases strongly depends on the interlayer materials and their thicknesses. The introduction of Al or Cu spacer layer does not provide any exchange coupling due to the interface instability. When the C interlayer thickness was about 0.5 nm, a strong interlayer exchange coupling between FePt and Fe has been realized and the saturation magnetization was largely improved. The high temperature magnetic properties of FePt/C/Fe films suggest that the stability of hard magnetic properties are improved by the interlayer exchange coupling, as compared to the bilayer FePt/Fe films. These interlayer exchange coupling seems to be a promising way in making exchange spring in future for the applications in permanent magnet.

Acknowledgements

This work was financially supported by the DRDO, New Delhi through a research project [ERIP/ER/0900363/M/01/1185]. Infrastructure facilities provided by DST, New Delhi [SR/S2/CMP-19/2006, SR/FST/PII-020/2009] are gratefully acknowledged.

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