JOURNAL OF OPTOELECTRONICS AND ADVANCED MATERIALS, Vol. 10, No. 12, December 2008, p. 3501 - 3504

The influence of the thickness layers and annealing conditions on the hard magnetic properties of nanocomposite [NdFeBNbCu/FeBSi]xn films

H. CHIRIAC, M. GRIGORAS, N. LUPU, M. URSE*

National Institute of Research & Development for Technical Physics, 47 Mangeron Blvd., 700050 Iasi, Romania

Some results concerning the influence of the thicknesses ratio of the NdFeBNbCu/FeBSi bilayers and annealing conditions on the structural and hard magnetic properties of the Ta/[NdFeBNbCu/FeBSi]·n/Ta multilayer thin films with low crystallization temperature are presented. The good hard magnetic characteristics were obtained for the Ta/[NdFeBNbCu(90)/FeBSi(5)]·6/Ta thin films annealed at 630°C for 20 min., and then cooled up down to RT in magnetic field. Thus, the Ta[NdFeBNbCu(90nm)/FeBSi(5nm)]·6/Ta thin films annealed for 20 min. at 630°C and then cooled up down to RT in magnetic field of 8 kG exhibit an increase in coercivity of about 1.37 kG and a noticeable enhancement of the saturation magnetization and the remanent magnetization of about 12 emu/g and 22 emu/g, respectively, as compared with same sample annealed at 630°C for 20 min., and then cooled up down to RT without magnetic field.

(Received November 7, 2008; accepted November 27, 2008)

Keywords: Thin films, Nanocomposite, Hard magnetic materials, Thickness layer, Multilayer, Annealing conditions, Soft and hard phases

1. Introduction

Nanocomposite magnets consisting of a fine mixture of magnetically hard and soft phases are very attractive materials for potential permanent magnet development [1]. These magnetic materials combine the advantages of both hard and soft phases [2, 3]. For hard magnetic materials thin films a lower crystallization temperature is desired for the reduction of the grain size and applications connected with Si technology [4]. The large values of the coercivity (Hc) and remanent magnetization (Mr) in exchange – coupled hard/soft nanocomposite multilayer films were achieved by the control of the thickness layers and annealing conditions. In this paper the influence of the thicknesses ratio of the NdFeBNbCu /FeBSi bilayers and annealing conditions on the structural and hard magnetic properties of the Ta/[NdFeBNbCu/FeBSi]·n/Ta multilayer thin films is reported. The aim of this study is the achievement of the hard magnetic thin films with low crystallization temperature.

2. Experimental details

Sandwich magnetic films of Ta/[NdFeBNbCu/FeBSi]·n/Ta have been prepared in vacuum by successive depositions using the r.f sputtering technique from three sputtering targets: a disc of $Nd_{12}Fe_{82}B_6$ alloy with Nb and Cu chips on its surface, a disc of $Fe_{70}B_{15}Si_{15}$ alloy and a disc of Ta. The samples were deposited, at room temperature, on silicium (100) substrates. The optimal composition for the NdFeBNbCu thin films was obtained by modifying the number of chips of Nb and Cu disposed on the surface of the target. All samples were sandwiched by two Ta layers with the thickness of 20 nm which were used as buffer and capping layers.

The crystallographic structure was investigated using X-ray diffraction (XRD) analysis. An X-ray diffractometer (Diffractometer D8 Advance) with a monochromatized Cu-Kα radiation was used, in a Bragg-Brentano arrangement.

The Warren-Averbach method [5] was used to estimate of the crystalline grain sizes (with an error of ± 15%).

The composition of the NdFeBNbCu layer was determined using the SEM/EDX technique.

The magnetic characteristics were determined by a vibrating sample magnetometer (VSM) at a maximum magnetic field of 30 kG applied parallel with the film plane.

The as-deposited samples were thermal treated in vacuum for 20 min. (optimum annealing time) [6] at temperatures between 630°C and 700°C. In order to improve the specific magnetic properties the selected samples were thermally treated in vacuum at temperature of 630°C and then it were cooled up down to room temperature (RT) in a magnetic field of about 8 kG applied in the film plane.

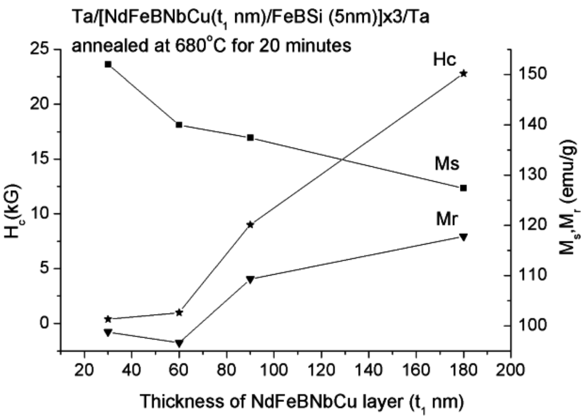


Fig. 1 The dependence of the coercivity, saturation magnetization M_s and remanence magnetization Mr on the thickness (t_1) of the NdFeBNbCu layer for the multilayer Ta/[NdFeBNbCu(t_1 nm)/FeBSi(5nm)] ·n/Ta films annealed at 680°C for 20 minutes

3. Results and discussion

Magnetic properties of Ta/[NdFeBNbCu(t_1)/FeBSi(t_2)]·n/Ta multilayer samples have been determined for films of various thickness of NdFeBNbCu layers from 30 to 180 nm and the FeBSi layers from 2.5 to 20 nm, while the total thickness of NdFeBNbCu layers was of 540 nm. Figure 1 shows the dependence of the coercivity Hc, saturation magnetization Ms and remanent magnetization Mr on the thickness (t_1) of the NdFeBNbCu layer for the Ta/[NdFeBNbCu($t_1$nm)/FeBSi (5nm)]·n/Ta multilayer films annealed at 680°C for 20 minutes. The annealing time of 20 minutes has been chosen to be the one which provided simultaneously the highest coercivity and Mr/Ms ratio values. In Fig.1 a large increase in the coercivity and a gradual decrease of the saturation magnetization Ms can be observed with the increase of the thickness of the NdFeBNbCu layers up to 180 nm. A small decrease of the remanent magnetization for the thickness of the NdFeBNbCu layer of about 30 nm and a gradual increase can be observed for thicknesses larger than 30 nm. The good results were obtained for two samples with ratios of the thickness NdFeBNbCu/FeBSi bilayers of 90/5 and 180/15.

The annealing temperature dependence of coercivity Hc and Mr/Ms ratio of Ta/[NdFeBNbCu (t_1 nm)/FeBSi (t_2 nm)]×n/Ta multilayer films (with t_1 = 90 nm and 180 nm and t_2 = 5 and 15 nm) is showed in Fig. 2. From Fig. 2 it can be seen that the best hard magnetic characteristics were obtained for Ta/[NdFeBNbCu(180nm) /FeBSi(15nm)]·3/Ta films annealed at 680°C for 20 min.

For many applications, compatible with Si technology, the thin film permanent magnets with low crystallization temperatures can be a good hard magnetic material for many applications. From Fig. 2 can be observed that the good hard magnetic values were obtained for Ta/ [NdFeBNbCu(90nm) /FeBSi(5nm)]·6/Ta thin films annealed at 630°C. A reduction of 50°C in the annealing temperature from 680°C to 630°C, when the hard/soft layers ratio increases from 12 at 18, leads for multilayer Ta/[NdFeBNbCu(90nm)/FeBSi(5nm)]·6/Ta thin films annealed at 630°C to an enhancement of the remanence Mr/Ms from 0.80 to 0.95 and good values for Hc of 17.1 kG.

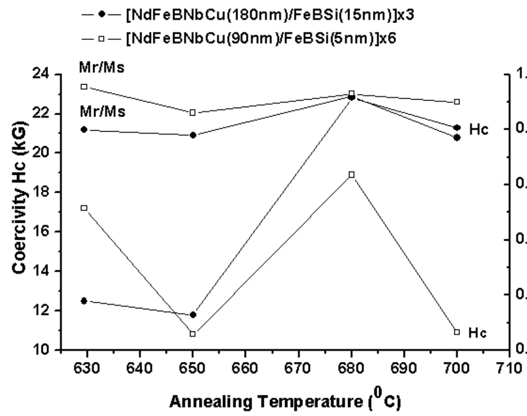


Fig. 2 The annealing temperature dependence of coercivity Hc and Mr/Ms ratio of multilayer Ta/[NdFeBNbCu(t_1)/FeBSi(t_2)] ·n/Ta films

XRD investigations show that samples in as – deposited state and after annealing at temperatures below 570°C have amorphous structure. At annealing temperatures between 570°C and 600°C the microstructure of the samples consists of a small number of Fe_3B nanograins embedded in the amorphous matrix. Samples annealed at temperatures higher than 600°C exhibit a complex multiphase structure of tens of nanometers. Figure 3 shows the XRD patterns for two selected samples: Ta/[NdFeBNbCu(90nm) /FeBSi(5nm)]·6/Ta thin film annealed at 630°C (curve a); Ta/[NdFeBNbCu(180nm)/FeBSi (15nm)]·3/Ta thin film annealed at 680°C (curve b). One can be observed that both samples present the same type of phases, but their volume is lower for Ta/[NdFeBNbCu(90nm) /FeBSi(5nm)]·6/Ta film annealed at 630°C. The addition of Cu and Nb is very effective in refining the $Fe_3B/Nd_2Fe_{14}B$ nanocomposite produced by crystallization.

Thus for the Ta/[NdFeBNbCu(90nm)/FeBSi(5nm)]·6/Ta film annealed at 630°C (curve a), the average crystalline size of $Nd_2Fe_{14}B$ phase is of about 22 nm while for Ta/[NdFeBNbCu(180nm) /FeBSi(15nm)]·3/Ta film annealed at 680°C (curve b) is about 31 nm. The average crystalline size of the soft magnetic phases for both samples is between 8 and 10 nm. The smaller thickness of hard and soft layers (90/5) leads to an enhancement of the diffusion process during the crystallization treatment at 630°C.

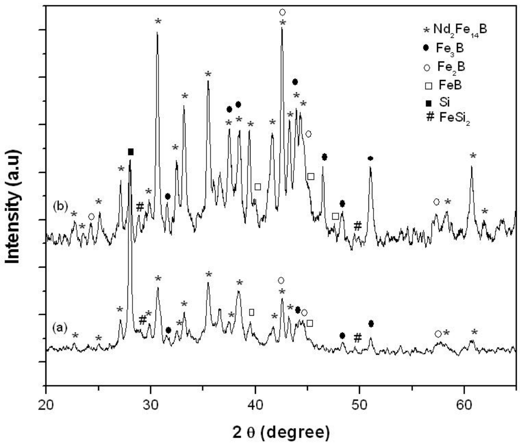
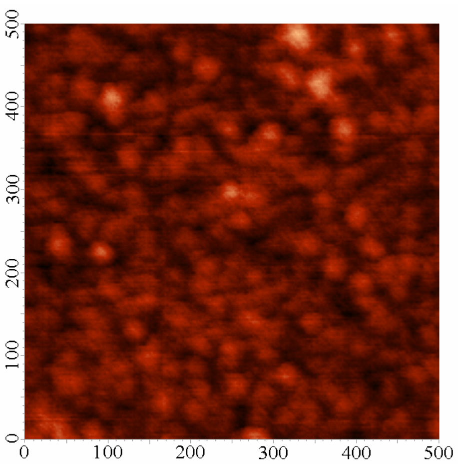
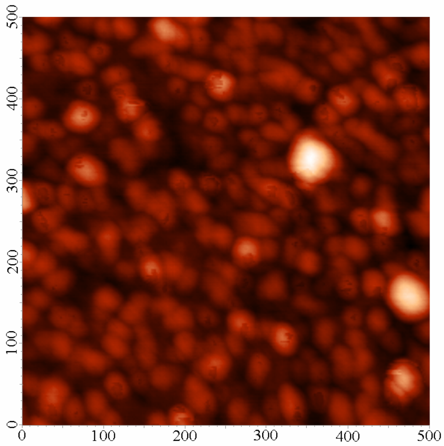


Fig. 3. XRD patterns for: [NdFeBNbCu(90nm)/FeBSi(5nm)] ·6 film annealed at 630°C for 20 min. (curve a); [NdFeBNbCu(180nm) /FeBSi(15nm)]·3 film annealed at 680 °C for 20 min. (curve b).



(a)



(b)

Fig. 4. AFM images for: (a) - Ta/[NdFeBNbCu(90nm)/FeBSi(5nm)]·6/Ta film annealed at 630°C, for 20 min.; (b) - Ta/[NdFeBNbCu(180nm)/ FeBSi(15nm)]·3/Ta film annealed at 680°C for 20 min.

In Fig. 4 (a, b) the AFM images of Ta/[NdFeBNbCu (90nm)/FeBSi(5nm)]·6/Ta thin film annealed at 630°C (curve a) and Ta/[NdFeBNbCu(180nm)/FeBSi (15nm)]·3/Ta film annealed at 680°C (curve b) are presented. The AFM image of the Ta/[NdFeBNbCu(90nm)/FeBSi (5nm)]·6/Ta film annealed at 630°C (curve a) presents a morphology consisting of irregular spheres particles. The average size of the particles is between 10 nm and 22 nm. The AFM image of the Ta/[NdFeBNbCu(180nm)/FeBSi(15nm)]·3/Ta film annealed at 680°C (curve b) presents a morphology consisting of regular spheres particles. The average size of the particles is between 18 nm and 29 nm. These AFM morphological results confirm the grain size values obtained by XRD technique.

The composition of Ta/NdFeBNbCu(50nm)/Ta single layer performed by SEM/EDX technique is follows: Fe 64.57 at.%, Cu 1.81 at.%, Nb 5.08 at.%, Nd 13.21 at.%, Ta 9.43 at.%, B up to 100 at.%. Figure 5 shows the δM plots for Ta/[NdFeBNbCu(180nm) /FeBSi(15nm)]·3/Ta thin films annealed at 680°C for 20 min., and Ta/[NdFeBNbCu(90nm)/FeBSi(5nm)]·6/Ta thin film annealed at 630°C for 20 min. For samples annealed at 680°C (curve a), only a positive *δM* is observed indicating the existence of the exchange coupling between the soft and hard grains. For samples annealed at 630°C (curve b) an initially negative δM for magnetic fields between 0 and about 14.8 kG was measured indicating the existence of the magnetostatic interactions between soft and hard grains. A positive *δM* is then observed indicating the existence of the exchange coupling between soft and hard grains.

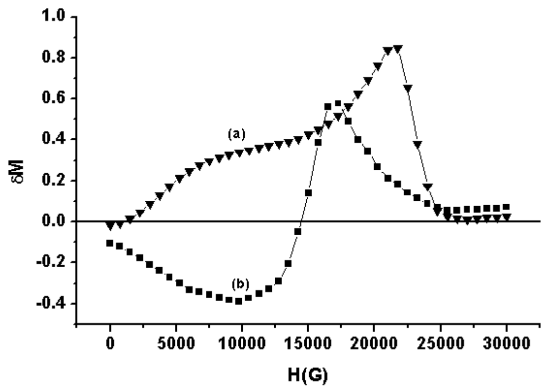


Fig. 5. dM versus applied magnetic field for: Ta/[NdFeBNbCu (90nm)/ FeBSi(5nm)]·6/Ta film annealed at 630°C (curve a); Ta/[NdFeBNbCu (180nm)/FeBSi(15nm)]·3/Ta film annealed at 680 °C (curve b).

In order to improve the hard magnetic properties of Ta/[NdFeBNbCu(90nm)/FeBSi(5nm)]·6/Ta thin films, the as-deposited samples were annealed at crystallization temperature and then were cooled up down to RT in magnetic field. Figure 6 shows the hysteresis loops of

Ta/[NdFeBNbCu(90nm)/ FeBSi(5nm)]·6/Ta sample annealed at 630^{o}C (loop a) and annealed at 630^{o}C, and then cooled up down to RT in magnetic field of about 8 kG (loop b).

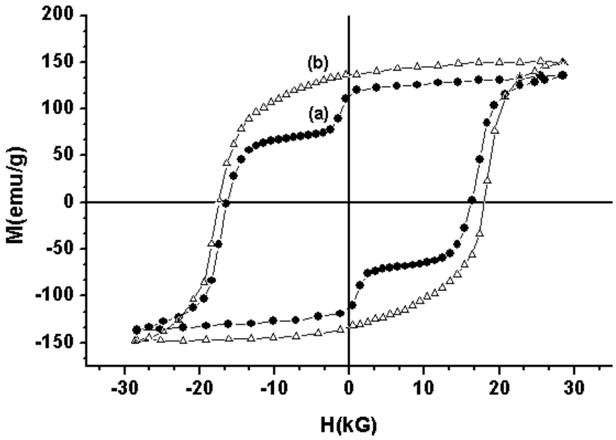


Fig. 6. Hysteresis loops for Ta/[NdFeBNbCu (90nm)/FeBSi(5nm)]·6/Ta film annealed at 630^{o}C (loop a), and then cooled up down to RT in magnetic field of 8 kG (loop b) .

It can be observed that the cooling in magnetic field leads to a noticeable enhancement of the saturation magnetization Ms of about 12 emu/g and remanence of about 22 emu/g. The exchange coupling is not improved significantly, fact confirmed by a low increase of the coercivity Hc of about 1.37 kG. In the same time the kink of the hysteresis loop from II quadrant disappeared and a better squareness was observed.

From the analysis of the presented data it results that although the Ta/[NdFeBNbCu(180nm)/ FeBSi(15nm)]·3/Ta thin films annealed at 680^{o}C for 20 minutes have the best hard magnetic properties, the Ta/[NdFeBNbCu(90)/FeBSi(5)]·6/Ta thin films annealed at 630^{o}C for 20 minutes and then cooled up down to RT in magnetic field can be a good hard magnetic material for applications connected with Si technology as micromagnets for micro-electromechanical systems (MEMS).

4. Conclusion

The hard magnetic properties of Ta/[NdFeBNbCu/FeBSi]·n /Ta multilayer thin films depend on the thicknesses ratio of the NdFeBNbCu/FeBSi bilayers and annealing conditions. For Ta/[NdFeBNbCu/FeBSi]·n/Ta thin films, an increase of hard/soft phases ratio from 12 at 18 and a reduction of annealing temperature of about 50^{o}C from 680^{o}C to 630^{o}C, lead to an enhancement of the remanence Mr/Ms from 0.80 to 0.95 and good values for Hc of about 17.1 kG. The Ta/[NdFeBNbCu(90nm)/FeBSi(5nm)]·6/Ta films annealed for 20 minutes at 630^{o}C and then cooled up down to RT in magnetic field of 8 kG exhibit a low increase in coercivity of about 1.37 kG and a noticeable enhancement of the saturation magnetization and remanence of about 12 emu/g and about 22 emu/g, respectively, as compared with same sample annealed at 630^{o}C for 20 min., and then cooled up down to RT without magnetic field.

Acknowledgment

This work was supported by the CEEX Excellency Research Program/ MATNANTECH Program (Contract no.19/2005) from the Ministry of Education, Research and Youth, ROMANIA and partial by a bilateral co-operation project ROMANIA – GREECE (Theme no.25).

References

[1] Chuan – Bing Rong, Vikas Nandwana, Narayan Poudyal, J.Ping Liu, Tetsuji Saito, Yaqiao Wu and Matthew J. Kramer, J. Appl. Phys., **101**, 9, 09K515 (2007).
[2] Chen, D.N. Brown, Bao Min Ma, P. Campbell, Y.Q.Wu, M.J.Kramer, IEEE Trans. Magn., **41**(10), 3864 (2005).
[3] Toshio Miyoshi, Hirokazu Kanekiyo, and Satoshi Hirosawa, IEEE Trans. Magn., 41, 10, .3865 (2005).
[4] Tsung-Shune Chin, Journal of Magnetism and Magnetic Materials, **209**, 1, 75 (2000).
[5] Rare- Earth Iron Permanent Magnets, Ed. by J. M. D.Coey, Clarendon Press, Oxford, (1996).
[6] H. Chiriac, M.Grigoras, N. Lupu, M.Urse and V. Buta, J. Appl. Phys., **103**, 07E144 - 3 pages (2008).

*Corresponding author: urse@phys-iasi.ro