Romanian Reports in Physics, Volume 56, No. 3, P. 352 - 358, 2004

Microstructure and magnetic properties in percolating [FeNiCoB/(SiO_2)]× n thin films

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Abstract. In this paper we report some experimental results concerning the influence of the composition, thickness of layers and annealing temperature on the magnetic, electrical and microstructural properties of [FeNiCoB/(SiO_2)]× n thin films. Close to the percolation threshold we have evidentiated the crucial changes in physical properties of these materials.

Key words: Thin films; Multilayers; Microstructure; Percolation threshold

1. Introduction

The metal – insulator films are composite materials consisting of metallic nanograins embedded in an insulating matrix. The nanogranular metal – insulator thin films display a wide variety of unusual physical properties, for values of the metal content, x, near a critical value, $x_{c,}$ usually called the “percolation threshold” [1,2]. The electrical and magnetic properties of metal – insulator films are strongly dependent on their composition and microstructure. The ferromagnetic metal – insulator thin films with very high electrical resistivity are an excelent candidate for high permeability cores in thin film inductors that can be used in IC (Integrated Circuit).

This paper reports some results concerning the influence of the FeNiCoB and SiO_2 layers thickness, layers composition and annealing temperature on the magnetic, electrical and microstructural properties of [FeNiCoB/(SiO_2)]×n thin films, close to the percolation limit. A comparison between the magnetic properties of FeNiCoB and [FeNiCoB/(SiO_2)]×n thin films is also reported.

2. Experimental Details

[FeNiCo/(SiO_2)]×n and [FeNiCoB/(SiO_2)]×n thin films were prepared using a conventional R.F. diode sputtering system (Laboratory Sputtering Plant Z – 400), by sequential deposition from two elemental targets: disc of FeNiCo alloy or disc of FeNiCo alloy with chips of B on their surface and disc of SiO_2. The samples with a thickness value of about 1,200 nm were deposited on various substrates depending on the intended measurements.

All the samples were thermally treated in vacuum at temperatures between at 300 and 400° C.

The structure of thin films was investigated using X-ray diffraction analysis. A X-ray diffractometer with a monochromatized Mo-Kα radiation was used, in a Bragg-Brentano arrangement. For X-ray diffraction analysis, [FeNiCoB/(SiO_2)]×n thin films were deposited on the molybdenum substrates.

The microstructure of the samples was investigated by transmission electron microscopy (TEM), using molybdenum 'microscope grids', coated with an evaporated carbon (8 – 10 nm) thin film as substrates. FeNiCoB/SiO_2 thin films with a total thickness of about 85 nm (65nm FeNiCoB/20nm SiO_2) were used for TEM analysis. The electron microscopy studies were carried out with a JEOL –200CX microscope.

The film composition was determined by electron microprobe analysis on [FeNiCoB/(SiO_2)]×n thin films deposited on molybdenum substrates.

The magnetic properties of [FeNiCo/(SiO_2)]×n and [FeNiCoB/(SiO_2)]×n thin films deposited on glass substrates were measured using a vibrating sample magnetometer in a magnetic field of up to 143 kA/m applied along to the film plane.

3. Results and Discussion

We studied the dependence of the resistivity of the [FeNiCoB/(SiO_2)]×n thin films on the values of the FeNiCoB/SiO_2 number layers (n). The thickness of FeNiCoB layer was varied from 60 to 360 nm, the thickness of SiO_2 was fixed at 40 nm and total thickness of [FeNiCoB/(SiO_2)]×n films was of about 1,200 nm.

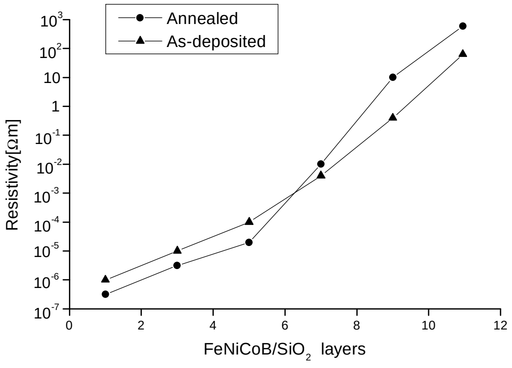

Fig. 1 - The dependence of the resistivity on the number layers (n) for [FeNiCoB/(SiO_2)]×n thin films

The dependence of resistivity on the value of the FeNiCoB/SiO_2 number layers in as-deposited and annealed samples (at 400^{o}C, for 2 h, in vacuum) is presented in Figure 1. The resistivity of [FeNiCoB/(SiO_2)]×n thin films is decreases exponentially by increasing the thickness of FeNiCoB layers within the multilayer system from 60 to 360 nm. After the thermal treatment at 400^{o}C, one can observe that a small increase in the resistivity values occurs for the samples with number layers higher than 6, whilst the samples with number layers below 6 present a decrease of resistivity values. The increase of the resistivity after the thermal treatment for samples with number layers higher than 6 can be associated with a diffusion process at the interface FeNiCoB/SiO_2.

The dependence of electrical resistance (ΔR/R) on temperature for [FeNiCoB/(SiO_2)]×n thin films, after annealing at 400^{o}C is presented in Figure 2.

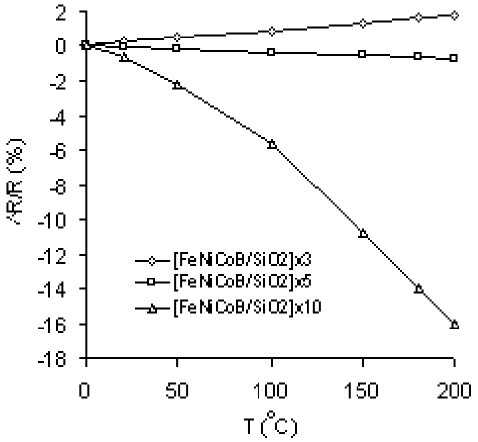

Fig. 2 - The dependence of relative electrical resistance (ΔR/R) on the temperature for [FeNiCoB/(SiO_2)]×n thin films

Fig. 2 reveals the influence of the number layers within multilayer on the electrical conduction mechanism. It can be see that the dependence of the resistance on temperature is typical for metallic materials in case of samples with the number layers below 5 and presents a non-linear dependence and negative values for ΔR in case of samples with the number layers higher than 5 and this suggests that the electrical conduction is determined by actived mechanisms.

In Figures 3 and 4 are presented the X-ray diffraction patterns for [FeNiCoB/(SiO_2)]×n thin films, in as – deposited state and after annealing at 400^{o}C, in vacuum, for 2 h.

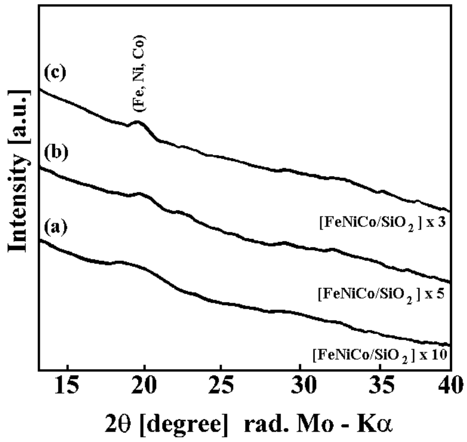

Fig. 3

X – ray diffraction patterns of as – deposited [FeNiCoB/(SiO$_2$)]×n thin films

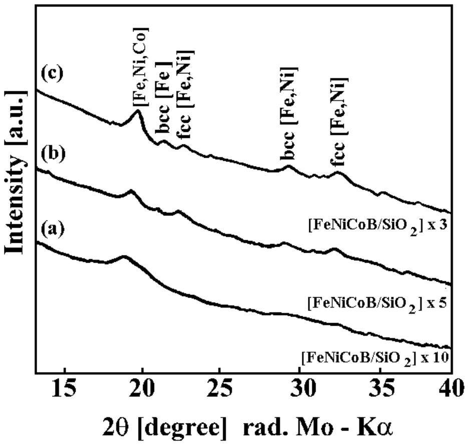

Fig. 4

X – ray diffraction patterns of annealed [FeNiCoB/(SiO$_2$)]×n thin films

The X-ray diffraction patterns of the as – deposited thin films samples show a nanocrystalline or an amorphous structure, as follows: nanocrystalline structure for the samples with the number layers n = 3 and n = 5 ($Fe_{55}Ni_{20}Co_{10}B_{15}$) alloy content, indicated by the small peaks; amorphous structure for samples with the number layers n = 10 ($Fe_{55}Ni_{20}Co_{10}B_{15}$) alloy content, indicated by a broad peak at 2θ = 19 deg., corresponding to the (Fe,Ni, Co) reflection. The effect of annealing on the microstructure was studied qualitatively. X-ray diffraction patterns corresponding to the [FeNiCoB/(SiO$_2$)]×n thin films after successively annealing in vacuum, at temperatures of 300^{o}C and 350^{o}C, for 2 h at each temperature, do not exhibit evident changes as compared to the X-ray diffraction patterns for as-deposited state, indicated that no major structural changes were produced by these treatments. After treating the [FeNiCoB/(SiO$_2$)]×n thin films at 400^{o}C, the X-ray diffraction patterns show changes in microstructure: the samples with the number layers n = 3 present a microcrystalline structure corresponding to a mixture of phases (FeNiCo) dispersed in the dielectric matrix; the samples with the number layers n = 5 and n = 10 present a nanocrystalline (FeNiCo) phase (the broad peak at 2θ = 19 deg.) dispersed in the dielectric matrix.

In Figure 5 is presented two electron micrographs, realized at 20^{o}C, for FeNiCoB (65 nm) thin film - Fig.5(a) and FeNiCoB(65 nm)/SiO$_2$(20 nm) thin film – Fig.5 (b).

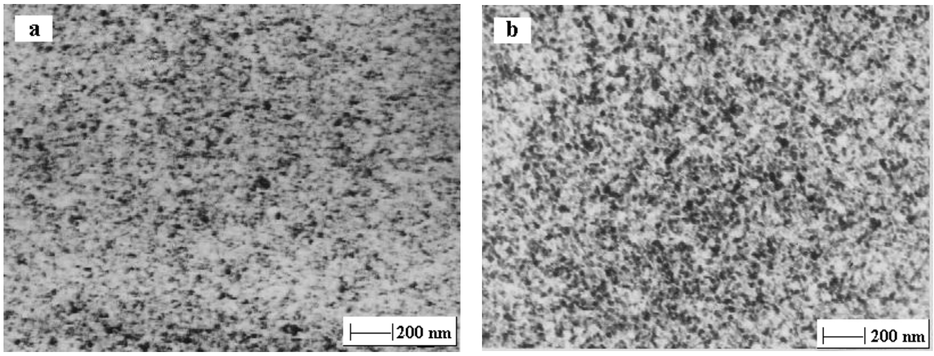

Fig.5 - Microstructure of the FeNiCoB thin films: (a) electron micrograph of the thin films; (b) electron micrograph of the FeNiCoB/(SiO_2) thin films

In generally, the microstructure of metal/insulator thin films consists of approximately spherical, very small metal particles separed by smaller barriers of insulator material. The electron micrograph for FeNiCoB (65 nm)/SiO_2 (20 nm) thin films shows a labyrinth structure consists by short filamentary chains with interconnected FeNiCoB particles and the dispersed insulator materials, filling in the free spaces. Most of FeNiCoB particles are discontinuous, but a significant fraction exists as small chains with bridges.

Fig.6 shows the hysteresis loops at room temperature for selected samples with the applied magnetic field parallel to the sample plane. In Fig. 6 one can see that the values of the saturation magnetization for FeNiCoB (1,000 nm) and [FeNiCoB(200 nm)/(SiO_2) (40 nm)]×5 thin films increase, whilst the coercive field values decrease, after annealing at 400^oC.

By anlyzing of the presented results can be see that the important changes in physical properties of [FeNiCoB/(SiO_2)]×n thin films occur by variation of the FeNiCoB/SiO_2 number layers (n). In Fig. 1 one can be see that the percolation point where resistivity verifies a huge upturn is presenting for a FeNiCoB/SiO_2 number layers, n = 5. In Fig. 2 one can see that FeNiCoB/SiO_2 number layers within the multilayer system have influence on the electrical conduction mechanism and thus it changes from the metallic behavior to activated mode.

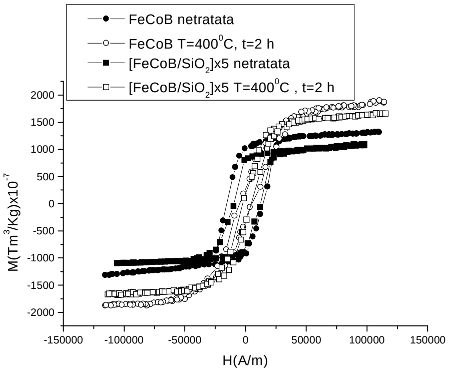

Fig. 6 - Magnetic hysteresis loops of FeNiCoB (1,200 nm) and [FeNiCoB (200 nm)/SiO$_2$ (40nm)]×5 thin films, in as-deposited and annealing state.

4. Conclusions

The [FeNiCoB/(SiO$_2$)]×5 thin films exhibit high resistivity over 10^{-5} Ωm, while superior soft magnetic properties, specific for FeNiCoB thin films are maintained. The annealed [FeNiCoB(200 nm)/(SiO$_2$)]×5 thin films show good magnetic properties of saturation magnetization ≅ $1570\cdot10^{-7}$ Tm^3/kg and coercivity ≅ 1990 A/m. The good electrical and soft magnetic properties of this thin films were due to the granular microstructure. This class of granular metal films may be interesting for electromagnetic micro-devices which can operating at high frequencies.

REFERENCES

1. D.BABONNEAU, F.PETROFF, J.-L. MAURICE, F.FETTAR, A. VAURES, Evidence for a self-organized growth in granular Co/Al_2O_3 multilayers, Appl. Phys. Lett. 76, 2892, 2000.
2. A.Y. DOVZHENKO, P.V.ZHIRKOW, The effect of particle size distribution on the formation of percolation clusters, Phys. Lett. A 204, 247, 1995.
3. P. SHENG, J. KLAFTER, Hopping conductivity in granular disodered systems, Phys. Rev. B 27, 2583, 1983.
4. P. SHENG, B. ABELES, Y. ARIE, Hopping conductivity in granular metals, Phys. Rev. Lett., 31, 44, 1973.