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MAGNETIC, ELECTRICAL AND STRUCTURAL PROPERTIES OF GRANULAR [FeCoB/(SiO_2)]$\times$ n THIN FILMS

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The granular metal – insulator films are composite materials consisting of metallic nanograins embedded in an insulating matrix. The ferromagnetic metal – insulator thin films with very high electrical resistivity are an excelent candidate for high permeability cores in thin film inductors which operate at high frequencies. A source of losses are eddy currents in the magnetic layers. A possible way to reduce these losses is to fabricate the magnetic/dielectric multilayers. This paper reports some results concerning the influence of the FeCoB and SiO_2 layers composition and annealing temperature on the resistive, magnetic and microstructural properties of [FeCoB/(SiO_2)]$\times$n multilayer thin films. As compared to the well known FeCoB alloys, widely used in the electromagnetic micro-devices which can operate at high frequencies, the [FeCoB/(SiO_2)]$\times$n ($n \geq 30$) thin films exhibit high resistivity over 8×10^{-3} m$\Omega\cdot$m while superior soft magnetic properties specific for FeCoB thin films are maintained. The [FeCoB/(SiO_2)]$\times$60 thin films after annealing at 300 ^oC show good resistive and magnetic properties of the resistivity $\rho \cong 49$ mΩ m, saturation magnetization Ms $\cong$ 149 emu/g and coercive field Hc $\cong$ 7 Oe.

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1. Introduction

In recent years, researches on magnetic granular thin films have been actively done with the aim of development of new materials with good magnetic properties and high resistivity. Soft magnetic nanogranular materials are subject of great interest because of their promising technological applications in field of the electromagnetic micro-devices, such as magnetic thin film inductors. The materials used for manufacturing magnetic thin films inductors need to present specific characteristics: high saturation magnetization (M_s), low coercivity (H_c), high effective permeability, low losses. A candidate for material exhibiting higher permeability and lower losses even in the GHz – order high – frequency region, could be nanogranular magnetic thin film. The nanogranular magnetic/dielectric thin films display a wide variety of unusual physical properties. Their electrical and magnetic properties are strongly dependent on their composition and microstructure. The hetero-amorphous structure of these thin films exhibits much higher resistivity in contrast to the conventional nanocrystalline structures because it holds magnetic softness without exchange interaction between the metal particles [1-3].

This paper reports some results concerning the influence of the FeCoB and SiO_2 layers thickness and annealing temperature on the electrical, magnetic and microstructural properties of [$Fe_{52}Co_{22}B_{26}$ /(SiO_2)]$\times$n multilayer thin films, in view of their utilization for manufacturing magnetic thin film inductors which can operate at high frequencies. A comparison between the resistive and magnetic properties of $Fe_{52}Co_{22}B_{26}$ and [$Fe_{52}Co_{22}B_{26}$ /(SiO_2)]$\times$n thin films is also reported.

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2. Experimental

The $[FeCoB/(SiO_2)]\times n$ multilayer thin films were prepared using a conventional R.F. diode sputtering system (Laboratory Sputtering Plant Z – 400), by sequential deposition in vacuum from elemental targets, disc of FeCo alloy with chips of B on their surface and disc of SiO_2, mounted on two separate guns. The samples composition was determined by electron probe analysis on FeCoB thin films deposited on the molybdenum substrates.

The $[FeCoB/(SiO_2)]\times n$ samples with a total thickness of FeCoB magnetic layers about 0.36 μm were deposited on various substrates, depending on the intended measurements. The $Fe_{52}Co_{22}B_{26}$ and SiO_2 thin films were obtained in following experimental conditions: input power of 200 W and 240 W respectively, room temperature for substrates, total pressure of argon (Ar) was about 2 Pa. For electrical and magnetic measurements the $[FeCoB/(SiO_2)]\times n$ multilayer thin films were deposited on glass substrates. The electrical resistivity of the samples was measured using the D.C. four – point probe technique and the magnetic characteristics (Ms and H_c) were measured using a vibrating sample magnetometer (VSM) in a magnetic field of 1.8 kOe.

The crystallographic structure was investigated using X-ray diffraction (XRD) analysis. An X-ray diffractometer with a monochromatized Co-Kα radiation was used, in a Bragg - Brentano arrangement. The microstructure was investigated with a transmission electron microscopy (TEM), using molybdenum 'microscope grids', coated with an evaporated carbon (8 – 10 nm) thin film as substrates. The $[FeCoB(12nm)/ SiO_2(2nm)]x5$ thin films with a total thickness of about 70 nm were used for TEM analysis. The electron microscopy studies were carried out with a JEOL –200CX microscope.

The structural and morphological analysis and electrical and magnetic measurements of the samples were made on as-deposited and thermally treated samples. The as-deposited FeCoB and $[FeCoB/SiO_2]\times n$ thin films were annealed at temperatures between 200°C and 400°C, for 2 h, in vacuum of 2×10^{-3} Pa.

3. Results and discussion

The resistive and magnetic properties of $[FeCoB/SiO_2]xn$ thin films strongly depend on composition and microstructure. The resistivity and magnetic characteristics of the $[FeCoB/SiO_2]xn$ multilayer thin films are very sensitive to FeCoB and SiO_2 layers thickness and microstructural changes caused by annealing. In our studies the thickness of FeCoB single layer was varied from 6 to 120 nm and of SiO_2 single layer from 1 to 20 nm, while total thickness of $[FeCoB/(SiO_2)]\times n$ multilayer films was of about 420 nm (360 nm for FeCoB layers and 60 nm for SiO_2 layers).

We studied the dependence of the resistivity on the $FeCoB/SiO_2$ number layers (n). Results concerning this dependence, for as-deposited and annealed FeCoB and $[FeCoB/SiO_2]\times n$ thin films (at 300 and 400 °C, for 2 h, in vacuum) are presented in Table 1.

Table 1. Values of the resistivity for as-deposited and annealed samples.

Samples	Resistivity (mΩ·m)		
	As-deposited	After annealing	
		300 °C	400 °C
FeCoB (360 nm)	2.2×10^{-3}	1.9×10^{-3}	0.2×10^{-3}
$[FeCoB(120\ nm)/SiO_2(20\ nm)]\times 3$*	-	3.2×10^{-3}	0.75×10^{-3}
$[FeCoB(60\ nm)/SiO_2(10\ nm)]\times 6$*	-	5.6×10^{-3}	1.3×10^{-3}
$[FeCoB(12\ nm)/SiO_2(2\ nm)]\times 30$	8.3×10^{-3}	8.1×10^{-3}	4.5×10^{-3}
$[FeCoB(6\ nm)/SiO_2(1\ nm)]\times 60$	48.9	48.2	25.6

* When the thickness of SiO_2 layers is large (> 7 nm), the resistivity of as-deposited samples can't be measured by D.C. four – point probe technique.

One can see that resistivity values for as-deposited [FeCoB/SiO_2] × n thin films increase with increasing the number layers (n), for the cases when the thickness of the SiO_2 layers is small (< 7 nm). The resistivity values for all the samples decreases after annealing at temperatures between 200 °C and 400 °C. A little slower decrease of the resistivity values can be noticed for the annealed samples at 300 °C. After annealing at 400 °C one can see that there is a sharp decrease of the resistivity values for all the samples. The decrease of the resistivity for the films annealed at temperatures up to 300°C is due to decrease of the number of structural defects in FeCoB layers induced in films during the deposition process. The sharp decrease of the resistivity of annealed thin films at temperatures higher than 300 °C, is due to the beginning of the recrystallization process in metallic layers, fact also suggested by TEM images. Is possible that the recrystallization process to can be associated with a diffusion process at FeCoB/SiO_2 interfaces.

The dependence of the magnetic characteristics (saturation magnetization M_s and coercive field H_c) on the FeCoB/SiO_2 number layers (n) is presented in Fig.1.

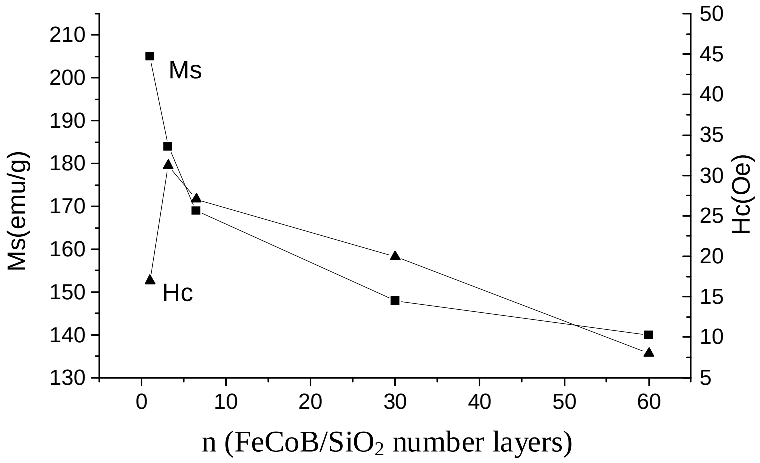


Fig. 1. Dependence of the magnetization and coercive field on the FeCoB/SiO_2 number layers (n).

One can see that the values of the saturation magnetization decreases with increasing the FeCoB/SiO_2 number layers while the values of the coercive field increase with increasing the FeCoB/SiO_2 number layers up to 3 and decrease afterwards.

In Table 2 are presented the values of the saturation magnetization and coercive field for as-deposited and annealed FeCoB and [FeCoB/SiO_2]xn thin films at temperatures of 300°C and 400°C.

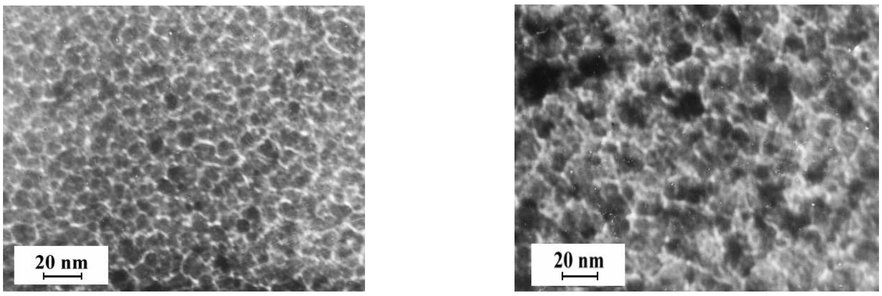
Table 2. Values of the saturation magnetization and coercive field for as-deposited and annealed samples.

Samples	Saturation magnetization (emu/g)			Coercive field (Oe)		
	As-deposited	After annealing 300 °C	After annealing 400 °C	As-deposited	After annealing 300 °C	After annealing 400 °C
FeCoB (360 nm)	205	208	215	17	18	25
[FeCoB(120 nm)/SiO_2(20 nm)]×3	184	187	195	32	30	56
[FeCoB(60 nm)/SiO_2(10 nm)]×6	169	172	198	27	25	69
[FeCoB(12 nm)/SiO_2(2 nm)]×30	148	154	203	22	18	71
[FeCoB(6 nm)/SiO_2(1 nm)]×60	140	149	211	8	7	89

In Table 2 one can see that after annealing at 300 °C, the small changes of the magnetic characteristics have occurred while after annealing at 400 °C the values of the saturation magnetization and coercive field undergoing significantly changes, namely the both values increase.

The structure of as-deposited and annealed FeCoB and [FeCoB/SiO_2]× n thin films was studied at room temperature with XRD and TEM techniques. All the XRD patterns present only small broadened peaks, possibly due to reduced size of randomly oriented (Fe, Co) crystallites. Estimation of the crystalline grain sizes was difficult because of the poor resolution of the diffraction peaks.

TEM images for as-deposited samples display nanometric spherical particles, with the average sizes are about 10 nm, embedded in a SiO_2 amorphous matrix, as seen in Fig. 2(a). Thermal treatments substantially alter the microstructure of nanogranular [FeCoB/(SiO_2)]×n thin films. In samples annealed at 400°C one can observe some larger particles, around 20 nm, as seen in Fig. 2(b).



(a) (b)

Fig. 2. TEM micrographs of [FeCoB(12nm)/ SiO_2(2nm)] × 5 thin films: in as-deposited state (a); after annealing at 400 °C.

By controlling the experimental conditions of preparation and layers thickness in multilayer systems, the nanogranular thin films with good resistive and magnetic properties were obtained.

4. Conclusions

In [FeCoB/(SiO_2)]× n sputtered thin films one can observe very different physical properties by changing the thickness for FeCoB and SiO_2 layers and after annealing at different temperatures. The [FeCoB/(SiO_2)]×n (n ≥ 30) thin films exhibit high resistivity over 8×10^{-3} mΩ·m while superior soft magnetic properties, specific for FeCoB thin films are maintained. The [FeCoB/(SiO_2)]× 60 thin films after annealing at 300 °C show good resistive and magnetic properties of the resistivity $\rho \cong 49$ mΩ·m, saturation magnetization Ms ≅ 149 emu/g and coercive field Hc ≅ 7 Oe. The good resistive and soft magnetic properties of these materials were due to the nanogranular microstructure.

The present results suggest that the [FeCoB/(SiO_2)] × n nanogranular thin films could be useful for electromagnetic micro-devices which can operate at high frequencies.

References

[1] K. Ikeda, K. Kobayashi, Fujimoto, J. Ceram. Soc. **85**, 169 (2002).
[2] X. N Jing, X. Yan, J. Appl. Phys. **83**, nr.11, part 2, 6530 (1998).
[3] M. Munakata, M. Motoyama, M. Motoyama, M. Yagi, T. Itoh, Y. Shimada, M. Yamaguchi, K. -I. Arai IEEE Trans. Mag. **38**, 3147 (2002).