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Deposition of Amorphous Fe-Zr Alloys by Magnetron Co-sputtering

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The formation of amorphous structure of Fe thin films by magnetron co-sputtering deposition dependence on Zr impurity at room temperature was explored. X-ray diffraction methods were applied for studying a amorphous phase forming range vs composition of elements. Electrical properties were evaluated by four-probe and van der Pauw methods. The surface morphology was investigated by SEM. It was found that amorphous structure of Fe-Zr metals alloy forms when Zr concentration is 9 at.%. The resistivity of amorphous and nanocrystalline $Fe_{1-x}Zr_x$ thin films is 2 – 3 times bigger than polycrystalline and slightly depends on composition.

Keywords: amorphous metals alloys, Fe-Zr alloys, FTS sputtering, magnetron co-sputering.

INTRODUCTION

During the last few decades intensive researches have been carried out on amorphous metal alloys because of their specific properties. Application of amorphous magnetic metallic alloys, also known as metallic glasses, plays an important role in computer industry, information technology, recording media, etc. [1 – 3]. These alloys are applied for spin dependent transport like giant magnetoresistance (GMR), tunneling magnetoresistance (TMR) devices, and nonvolatile memory (MRAM) [4]. Very smooth interface with small-scale roughness among the layers is necessary for realizing the high MR (magnetoresistance) values in sandwich GMR and TMR structures. Detailed studies have shown that GMR properties are extremely sensitive to the atomic scale structure of the interface. A significant decrease in the GMR ratio occurs when the average spacing between the ferromagnetic layers is changed, even by as little as 2 Å [5]. In addition, if the layers have a roughness comparable in amplitude and wavelength to the spacer layer thickness, Neel coupling occurs and it becomes more difficult to switch the magnetic moment of the layers. Contamination of a layer by metal atoms of another layer causes an increase in spin independent scattering [6] and a loss of local magnetic alignment [7]. Therefore atomically smooth, unmixed interfaces are necessary [8, 9]. Knowledge on influence of processing conditions on atomic scale defects is insufficient. So, the current deposition processes have not produced theoretically predicted GMR properties. The way to solve these problems is to use amorphous magnetic materials with high quality of surface. To obtain amorphous metal alloys there are several available techniques. They can be divided into two groups: metallurgical and micro technological (deposited by sputtering or evaporation, solid-state reactions, ion beam mixing, layer-by-layer deposition) [10 – 14].

This paper introduces experimental analysis of $Fe_{1-x}Zr_x$ alloys production by magnetron co-sputtering of Fe and Zr metals on glass substrate. The aim of the work was to determine the minimum concentration of Zr impurities enabling formation of amorphous structure of Fe thin films as well as to investigate electrical and morphological properties of these films.

EXPERIMENTAL

Various compositions of metal alloys $Fe_{1-x}Zr_x$ ($x = 0 – 1$) coatings were obtained by magnetron sputter co-deposition. (Fig. 1). $Fe_{1-x}Zr_x$ films were deposited on glass substrates by facing target sputtering (FTS) using magnetron (Fe-target) [15 – 16] and conventional magnetron (Zr target). The distance of Zr target to substrate and the distance of central axis of the target to substrate were 190 mm and 100 mm, respectively.

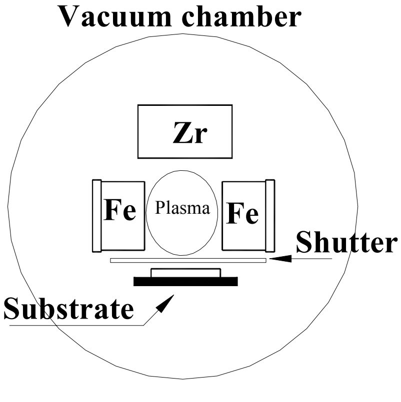

Fig. 1. Magnetron sputter co-deposition scheme (view from the top)

The 50 m^3/h rotary pump and 500 l/s diffusion pump were used to produce a vacuum. Residual gas pressure was $5\cdot10^{-4}$ Pa. Before deposition glass substrates were cleaned using standard procedures of microelectronics technology. After the chemical treatment all substrates were cleaned using oxygen plasma processing. Other deposition parameters are presented in Table 1. Target voltage and

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discharge current were measured using digital multimeters. Deposition rate was monitored by quartz crystal monitor and by weighing the films before and after deposition with the GR-202 balance having a resolution of 0.01 mg. X-ray diffraction (Cu-Kα radiation, $\lambda = 0.15405$ nm, Breg-Brentano geometry) was applied to define the structure of various composition of $Fe_{1-x}Zr_x$ coatings deposited on the substrates at room temperature.

Four-probe and van der Pauw methods were used to measure the electrical conductivity and to follow possible structure and phase composition transitions. The average size of crystallites of thin films was determined from the peak broadening by single line and multiple line analysis [17] using the program WinFit and XFIT.

Table 1. Films preparation conditions

Parameter	Fe	Zr
	FTS-magnetron	Conventional magnetron
Deposition temperature, ºC	20	
Ar gas pressure, Pa	0.7	
Deposition rate, mg/min·cm^2	0.16 – 0.64	
Thickness of film, μm	0.1 – 2	
P, W	1200	425 – 1800

ANALYSIS AND RESULTS

The X-ray diffraction patterns of $Fe_{1-x}Zr_x$ (x = 0.03, x = 0.05 and x = 0.09) films deposited on glass substrates at room temperature are shown in (Fig. 2). It shows that polycrystalline (Fig. 2, a, b, c) and amorphous coatings (Fig. 2, d) were obtained depending on the composition of the $Fe_{1-x}Zr_x$. Table 2 summarizes the main results of the XRD analysis: it gives values of diffraction angle 2θ, interplanar distance, and grain size. It should be noted that grain size decreases with increasing Zr concentration.

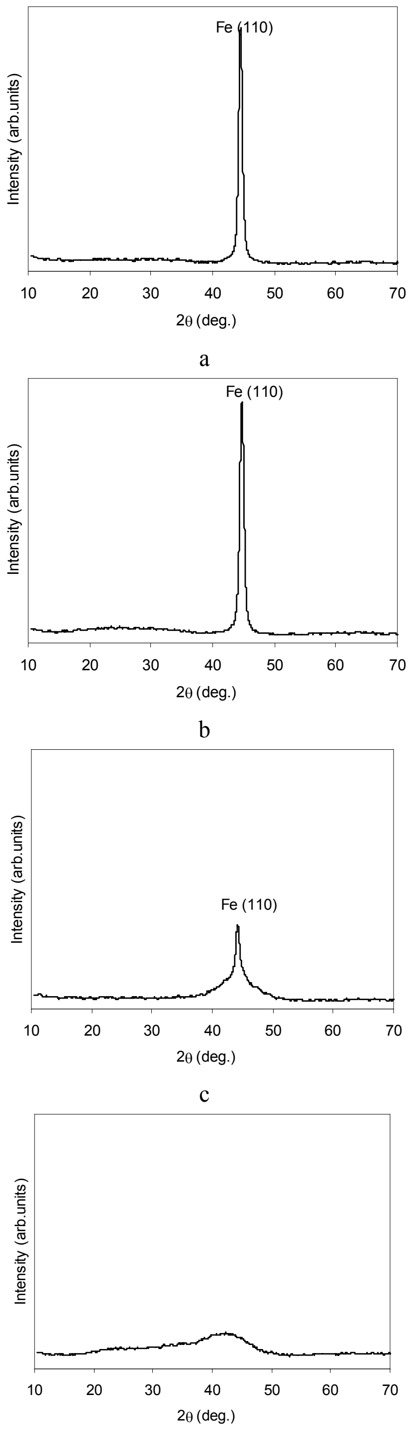
Concentration of elements was determined by Vegard's formula (for crystalline materials dependence of interplanar distance on concentration of elements):

$$a = a_A C_A + a_B C_B\,. \tag{1}$$

where a is the interplanar distance, a_A, a_B are the interplanar distance of component determined from XRD diagrams, C_A, C_B are concentration of components. Fe (110) shift in peak position is consistent with Vegard's law which states that the lattice parameter of a binary solid solution is proportional to the atomic percentage of the alloy (Fig. 2).

Table 2. XRD analysis results

	2Θ, deg	Interplanar distance, nm	Crystallite size, nm	Structure
α-Fe	44.487	0.2036	24	crystal
$Fe_{0.97}Zr_{0.03}$	44.268	0.2044	21	crystal
$Fe_{0.95}Zr_{0.05}$	43.913	0.2060	20	crystal
$Fe_{0.91}Zr_{0.09}$	42.174	0.2161	1.5	amorph.



a

b

c

d

Fig. 2. XRD patterns for $Fe_{1-x}Zr_x$ alloy thin films on glass substrate at various compositions: a – Fe; b – $Fe_{0.97}Zr_{0.03}$; c – $Fe_{0.95}Zr_{0.05}$; d – $Fe_{0.91}Zr_{0.09}$

Widths of the peak increases that indicates decrease of the crystallite size. $Fe_{0.91}Zr_{0.09}$ composed thin films show that amorphous structure is formed. $Fe_{0.91}Zr_{0.09}$ alloy could be considered as amorphous because the average crystallite size was 1.5 nm. Usually thin films with crystallite size less than 2.5 nm are called amorphous.

Resistivity of $Fe_{1-x}Zr_x$ alloys increases with concentration of Zr from $7.7\cdot10^{-7}$ Ω·m in ($Fe_{0.97}Zr_{0.03}$) to $14.8\cdot10^{-7}$ Ω·m in ($Fe_{0.95}Zr_{0.05}$) and $17.8\cdot10^{-7}$ Ω·m in ($Fe_{0.91}Zr_{0.09}$) in accordance with:

$$\rho_{\mathrm{Fe}_{1-x}\mathrm{Zr}_x} = \rho_{\mathrm{Fe}} + \alpha C_{\mathrm{Zr}} \cdot \qquad (2)$$

where ρ is the resistivity of alloy, C_{Zr} is concentration of component; α is linearity coefficient, ρ_{Fe} the resistivity of a pure Fe thin film.

The resistivity of amorphous binary metallic thin films is larger than polycrystalline films. The coefficient α is found to be $17 \cdot 10^{-6}$ Ω·m. This dependence and coefficient α is similar to the other metal-metal alloys [18, 19].

SEM pictures show rough surface in crystalline Fe thin film and smooth surface of amorphous $\mathrm{Fe}_{0.91}\mathrm{Zr}_{0.09}$ alloy.

CONCLUSIONS

Fe layers resistivity dependence on concentration of Zr impurity and crystal state was investigated. It was found that amorphous Fe-Zr metals alloy is formed by magnetron sputtering co-deposition when Zr concentration is 9 at.% at room temperature substrate. Resistivity of the amorphous and crystalline $\mathrm{Fe}_{1-x}\mathrm{Zr}_x$ thin films is 2 – 3 times bigger than the resistivity of polycrystalline films. Only slight dependence of resistivity on composition was observed. It was found that crystallite size decrease correlates with the surface structure of thin films that becomes smoother.

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