Available online at www.sciencedirect.com
ScienceDirect
Journal of Magnetism and Magnetic Materials 308 (2007) 101–107
Journal of magnetism and magnetic materials
www.elsevier.com/locate/jmmm

Influence of heat treatment on the structure and magnetic properties of the Co/Zr and Co/Hf multilayer amorphous films

S.S. Grabchikov*, L.B. Sosnovskaya, K.I. Yanushkevich

United Institute of Solid State Physics and Semiconductors of the Academy of Science of Belarus, P. Brovki street, 17, 220072 Minsk, Belarus

Received 22 February 2005; received in revised form 6 February 2006
Available online 5 June 2006

Abstract

The influence of annealing on the structure and magnetic properties of amorphous Co/Zr and Co/Hf multilayer films was studied with particular attention to the dependence of the magnetic properties, thermal stability and crystallization process on layer composition and thickness. The temperature at which crystallization commences increases from 400 to 460 °C as the layer thickness d_{Zr} or d_{Hf} increases from 6 to 18 Å, and decreases from 450 to 400 °C as d_{Co} increases from 12 to 18 Å. Multilayers containing 19–60 at% Zr were studied. The specific magnetization was found to increase even below the temperature at which crystallization commences. Our data are compared with non-multilayer Co–Zr amorphous films and rapidly quenched metallic glasses.

PACS: 75.70.−i

Keywords: Electrodeposition; Multilayer; Amorphous alloy; Magnetic properties; Heat treatment

1. Introduction

In recent years amorphous Co–TM (transition metal) alloy films with TM being Zr [1–3], Ti [2,3] or Nb [3], for example, have been much investigated because they have better magnetic characteristics and higher thermal stability than TM–metalloid amorphous alloys. Their great promise for use in thin film heads for high-density magnetic recording has been reported in several papers [4,5].

Single-layer TM–TM amorphous films can be sputter-deposited using composite targets. However, while such films have good magnetic characteristics, their structure is insufficiently homogeneous on the submicron level [6,7]. Earlier works [8,9] showed that although amorphous multilayer films (compositionally modulated structures [10]) are chemically inhomogeneous on the nm-scale they possess much higher structural homogeneity in the range 100–1000 Å than non-multilayer films.

Very interesting results were obtained during an investigation of the temperature dependence of the magnetization (J_s) of amorphous single-layer $Co_{88.1}Zr_{11.9}$ and multilayer CoZr/Si [11] and Co/Ti [12] films. For amorphous $Co_{88.1}Zr_{11.9}$ films J_s decreased with increasing temperature T, but in the range $T\sim600$ °C it increased at the beginning of the process of crystallization. For multilayered CoZr/Si films the practically linear dependence of J_s on T characteristic of two-dimensional ferromagnetswas observed [11]. For multilayered Co/Ti films there were two maxima on the $J_s(T)$ curve at 380 and 500 °C, corresponding to the crystallization of Ti and Co.

In the present paper, the structure and magnetic properties of amorphous Co/Zr and Co/Hf multilayer films are investigated together with the influence of thermal treatment.

2. Experimental

Amorphous Co/Zr and Co/Hf multilayer films containing 19–60 at% Zr or Hf were prepared by ion-plasma sputtering as described in Ref. [8]. The thickness of the individual Co (d_{Co}), Zr (d_{Zr}) and Hf (d_{Hf}) layers was measured using a Taylor–Hobson stylus profilometer and a MII-4 optical interferometer. The thickness d_{Co} was in the

*Corresponding author. Tel./fax: +375 017 284 11 28
E-mail address: gss@ifttp.bas-net.by (S.S. Grabchikov).

0304-8853/$ - see front matter
doi:10.1016/j.jmmm.2006.05.008

range 2–24 Å, d_{Zr} and d_{Hf} were in the range 2–18 Å and the total film thickness was 3000–5000 Å. We varied the number of repeats between 60 and 1250.

The chemical composition of the multilayers (weight %) was calculated according to

$$\mathbf{M}_{Co}(\mathrm{wt\%}) = (1 + p_{Zr}/p_{Co} d_{Zr}/d_{Co})^{-1}, \quad (1)$$

where p_{Zr} (or p_{Hf} for the Co/Hf system) and p_{Co} represent the density of the corresponding elements. To convert from the wt% to at% we used (2) [13]:

$$M_{Co}(\mathrm{at\%}) = M_{Co}(\mathrm{wt\%}) A_{Co} \left[M_{Co}(\mathrm{wt\%})/A_{Co} + M_{Zr}(\mathrm{wt\%})/A_{Zr} \right]^{-1}, \quad (2)$$

where A_{Co} and A_{Zr} are the atomic masses of the corresponding elements. The dependence of composition on the individual layer thicknesses is shown in Fig. 1.

Film structures were characterized by X-ray diffraction using Co–K_{α} radiation and by transmission electron microscopy (TEM). For TEM investigations we used Co/Zr films with thickness about 500–700 Å, separated from the substrates. For TEM investigations of individual Co layers we used Co films with thickness 18, 54 and 72 Å, deposited on to amorphous carbon layers.

The films were isothermally annealed for 2 h in vacuum (better than 3×10^{-5} Torr) in the range of 100–600 °C. After annealing, magnetic properties were studied at room temperature and the film structure was investigated.

Magnetic properties (coercive field H_C, magnetic anisotropy field H_k, squareness of the hysteresis loop M_r/M_S) were determined at room temperature using a *B–H* looper operating in fields up to 100 Oe. The saturation magnetic moment per unit mass (σ_s) was measured as a function of temperature in a field of 8.6 kOe applied in the plane of the sample (area $5 \times 5\,\mathrm{mm}^2$) using a Faraday balance [14]. The temperature range was $T = -196$ to 927 °C and the heating/cooling rate 200–250 °C/h. During the $\sigma_s(T)$ measurements, the samples were maintained in vacuum.

3. Results and discussion

The amorphous Co/Zr and Co/Hf multilayer films possessed magnetic anisotropy with the easy-axis direction in the film plane. As-deposited films were characterized by the following magnetic parameters: $H_C = 0.02$–0.08 Oe; $H_k = 6$–18 Oe: $M_r/M_S = 0.95$–0.98. The magnetic properties of Co/Zr films as a function of the composition and individual layer thicknesses were investigated previously [8].

We used X-ray structural measurements and the value of H_C, which is known to be highly sensitive to the presence of crystalline order [15], to monitor the effects of annealing. Fig. 2 gives H_C (T) data for Co/Zr films having constant $d_{Co} = 12$ Å and varying d_{Zr}. For the (12 Å Co/6 Å Zr) multilayer (curve 1 in Fig. 2) H_C starts to change after annealing at $T = 400$ °C. We denote this temperature as $T_{on.crys}$ (the temperature for the onset of crystallization). For larger d_{Zr} an increase in H_C is observed at higher temperatures 430–460 °C (curves 2 and 3). The increase in thermal stability as d_{Zr} increases from 6 to 18 Å is confirmed by the X-ray structural analysis data.

Fig. 3 shows X-ray diffraction patterns for Co/Zr multilayers containing 28 at% Zr as-deposited (1) and after annealing at $T = 420$ (2), 480 (3) and 600 °C (4). As seen from Fig. 3, the first crystalline diffraction peaks observed correspond to the (5 1 1) and (4 4 0) planes of the $Co_{23}Zr_6$ intermetallic compound. The intensity and number of crystalline diffraction peaks increases with annealing temperature. All the peaks observed on completion of the crystallization process for this film correspond to $Co_{23}Zr_6$ (Fig. 3, curve 4). For the films with 19 and 24 at% Zr a solid solution of a phase based on HCP Co (α-Co) and $Co_{23}Zr_6$ is observed, while for Zr concentration up to 32 at% we find a mixture of $Co_{23}Zr_6$ and Co_2Zr. Quantitative data are presented in Table 1.

Our X-ray structural studies show that the transition from the amorphous to the crystalline state may be

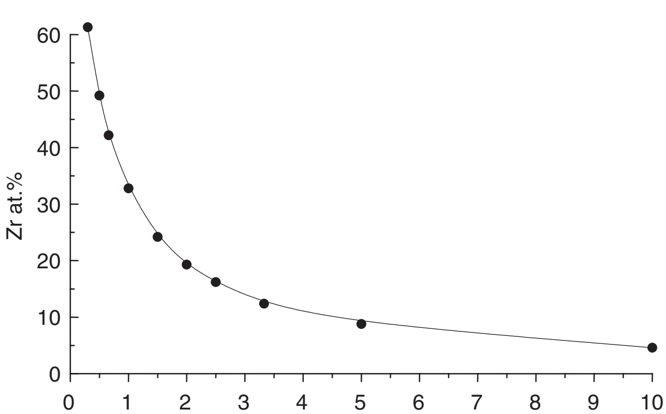

Fig. 1. Chemical composition of the Co/Zr multilayers as a function of the ratio of the Co and Zr layer thicknesses.

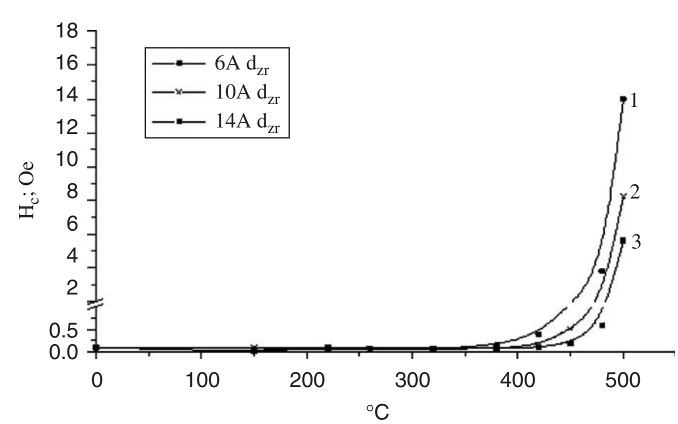

Fig. 2. Coercivity as a function of temperature for amorphous Co/Zr multilayer films with $d_{Co} = 12$ Å and $d_{Zr} = 6$ Å (1); 10 Å (2); 14 Å (3) i.e. 19 at% Zr; 28 at% Zr; 34 at% Zr.