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A Study of Interdiffusion Phenomena in Fe/Co-Nb-Zr Multilayers

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Abstract : R.f. sputtered Fe/CoNbZr multilayers are studied by means of conversion electron Mössbauer spectroscopy (CEMS) and X-ray diffraction (XRD). In the as-deposited conditions, an interdiffusion layer thickness of about 1.3 nm can be calculated from the CEMS data. Subsequently the samples were annealed in vacuum for 2h at 570 K. It is then estimated the percentage of interfacial Fe alloy formed, the structure of which is very likely nanocrystalline.

1. INTRODUCTION

High information density video recording requires heads possessing core materials with a large saturation magnetization, a high permeability and a magnetostriction constant close to zero. The first requirement can be fulfilled with Fe-rich alloys, the second one by obtaining the Fe grains as small as possible. Such a material can be prepared by inducing repeated nucleation of Fe grains on an intermediate layer. Recently good soft magnetic properties have been reported for Fe/Co-Nb-Zr multilayers by Dime et al. [1]. Here the basic idea is to utilize the high magnetization of Fe layers while the grain growth is controlled by interrupting it with an amorphous Co-Nb-Zr layer. It is also clear that the interfacial alloy formed by interdiffusion during either preparation or further annealing would play a key role in determining the final properties. In a recent paper, the influence of the interdiffusion layer on the magnetostriction constant was reported [2]. Therefore it is essential to get maximum information on the mixing at the interfaces. Conversion electron Mössbauer spectroscopy (CEMS) has proved to be a good technique for studying interface phenomena in the case of Fe/Co [3] and Fe/Zr [4] multilayers.

We prepared Fe/amorphous Co-Nb-Zr multilayers by sputtering and investigated the interdiffusion phenomena using the CEMS technique, which we report in this paper. The crystalline or amorphous state of the layers was also checked by means of X-ray diffraction (XRD) experiments. The studies were first conducted in the as-deposited conditions and then after further annealing of the samples. Preliminary results were already published in Ref. [5].

2. EXPERIMENTAL

The multilayered samples were deposited by sequential r.f. sputtering using a chamber which was first pumped to a pressure of 4×10^{-7} Torr. The targets were pure Fe (99.99%) and an amorphous alloy of Co-Nb-Zr (CNZ). In order to improve the sensitivity of CEMS studies, a small chip of ^{57}Fe was placed on the target. The deposition parameters were r.f. power 80 W and argon pressure 6 mTorr. Water cooled glass substrates were used. The deposition rate was measured with a quartz crystal monitor which was first calibrated against the thickness obtained from profilometer measurements on thicker layers. The main features of the prepared samples (A, B, C, D) as listed in table 1. The actual composition of the CNZ layers was determined by electron microprobe analysis and was found to be $\text{Co}_{0.8}\text{Nb}_{0.1}\text{Zr}_{0.1}$ which was also close to that of the target. We chose this composition because earlier studies showed that the magnetostriction for this was 1.7×10^{-7} which is indeed very small [6].

CEM spectra were recorded by using a constant acceleration Mössbauer set up with $^{57}\text{Co}:\text{Rh}$ (50 mCi) as a source. A gas flow ($\text{He}+5\% \text{CH}_4$) proportional counter [7] was used to register the emitted conversion and Auger electrons subsequent to the resonant γ -ray scattering by ^{57}Fe nuclei. All spectra were recorded at room temperature and then fitted by standard procedures to obtain hyperfine interaction parameters. All isomer shifts δ are quoted with respect to that of $\alpha\text{-Fe}$.

XRD experiments were performed at LURE (Laboratoire pour l'Utilisation du Rayonnement Electromagnétique, Orsay, France) using a home-made set up especially adapted for thin film analysis [8]. It was designed for working with an intense X-ray source and allowed the spectra to be recorded with a position sensitive detector. Both incident and diffracted X-ray paths were placed under vacuum in order to optimize the signal to noise ratio. The wavelength was taken as 0.2254 nm in order to avoid K-Fe fluorescence.

Table 1 : Main features of the multilayered samples ($t(\text{Fe})$ = thickness of Fe layers, $t(\text{CNZ})$ = thickness of Co-Nb-Zr layers, n = number of bi-layers).

Sample	$t(\text{Fe})(\text{nm})$	$t(\text{CNZ})(\text{nm})$	n
A	12	2.4	6
B	6	2.4	10
C	2.4	2.4	16
D	2.4	4.8	15

3. RESULTS AND DISCUSSION

3.1. As-deposited conditions

Representative CEM spectra taken in the as-deposited conditions are presented in Fig.1a,b. The spectra obtained from samples A and B, i.e. those having the thickest Fe layers, are very similar, hence only the spectrum of sample A is shown in Fig.1a. It exhibits a six-line pattern which corresponds to crystalline $\alpha\text{-Fe}$ with an hyperfine field of 33 T. The relative intensities of the lines are close to 3:4:1:1:4:3 and hence show that the magnetization is in the film plane, as expected from magnetic measurements [5]. Only the spectral lines are slightly enlarged with respect to the standard Fe. Hence the results concerning both samples A and B indicate that the initial mixing at the interfaces is negligible; only 10%, or less, of the Fe atoms are concerned. On the contrary the spectra taken from samples C and D, i.e. those having the thinnest Fe layers (2.4 nm), are different from the previous one and indicate notable mixing at the interfaces. Fig.1b shows the typical spectrum for sample C and one observes an increase in linewidth which is more evident at the base of the lines. The latter spectrum can be resolved using the following set of spectral components :

Figure 1 : CEM fitted spectra taken from as-deposited multilayers : a) sample A; b) sample C.

(i) a sextet S_1 attributed to unmixed Fe atoms; (ii) a second sextet S_2 due to Fe with Co as first neighbours : this configuration leads to increase in both the hyperfine field and the Fe moment [9]; (iii) a third sextet S_3 arising from Fe with Zr and/or Nb as first neighbours : this configuration leads to reduction in the hyperfine field [10]; the formation of both Fe-Zr and Fe-Nb bonds is probable since the theoretical formation enthalpies for FeZr and FeNb equiatomic alloys are -36 and -25 kJ/mole, respectively [11]. The results of the fitting are listed in table 2 for sample C; they are also nearly the same for sample D.

Table 2 : Summary of CEMS results obtained for as-deposited sample C : H = hyperfine field, δ = isomer shift, W = linewidth, RI = relative intensity. The subspectra S_1 , S_2 , S_3 are defined in the text.

Subspectrum	H(T)	δ (mm/s)	W (mm/s)	RI (%)
S_1	33	0	0.26	46
S_2	35.5	0.02	0.44	35
S_3	31.1	-0.07	0.45	19

The main conclusion is that, from the relative intensity of both S_2 and S_3 subspectra, one can estimate that 54% of the Fe is alloyed. In other words, a 1.3 nm thick Fe layer is getting intermixed; the interdiffusion zone likely remains crystalline. Similar result was already pointed out in Ref. [1] and for Fe/Co multilayers as well [3]. Otherwise the obtained hyperfine interaction parameters are in agreement with those available in the literature for bulk iron-based alloys. The hyperfine field value of 35.5 T calculated for subspectrum S_2 (Fe-Co bonds) is consistent with that for bcc FeCo alloys with ~ 15 at.% Co [9]. Similarly, the isomer shift value for subspectrum S_3 (Fe-Zr and Fe-Nb bonds) is negative as expected [10,12].

3.2. Annealing process

Annealing was carried out at 570 K for 2h in a vacuum system with a base pressure better than 1×10^{-6} Torr. As in section 3.1, the results are close to each other for the first two (A,B) and the last two (C,D) samples. Representative CEM spectra are shown in Fig.2a,b and one observes, in addition to the initial six lines, a broadened component attributed to either a magnetic amorphous phase or a superparamagnetic nanocrystalline structure; the corresponding background curvature is more pronounced in Fig.2b than in Fig.2a. The spectra can be resolved by using the same set of subspectra (S_1 , S_2 , S_3) as previously plus a distribution of hyperfine field in the range 0-40T in order to take into account the broad component. Under these conditions, the fraction of mixed Fe in sample B is found to be 36%, including Fe in subspectra S_2 , S_3 9% and Fe in field distribution 27%. The thickness of the unmixed Fe layers then drops from 6 to 3.8 nm. Similarly for sample C, the same three Fe percentages are 78%, 27% and 51% respectively. The average hyperfine field value taken from the distribution is around 15T. The thickness of unmixed Fe layers does not exceed now 2-3 interatomic distances. It is also worth noticing that, for the four annealed samples, the relative intensity of the resonance lines supports preferential in-plane magnetic anisotropy. In summary,

Figure 2 : CEM fitted spectra taken from multilayers annealed for 2h at 570 K : a) sample B; b) sample C.

Figure 3 : High-angle X-ray diffractograms for samples B, C, D annealed at 570 K for 2h.

the CEMS study shows that the annealing process is not a simple continuation of the interdiffusion phenomena observed in the as-deposited conditions. It mainly points out a new broad component characteristic of a nanocrystalline or amorphous structure.

The structure of the annealed multilayers was further investigated by means of XRD. The scans were performed for $40^\circ < 2\theta < 140^\circ$ and diffraction peaks were only detected in the range $64-70^\circ$ which also includes the position of the standard Fe (110) line. The high-angle X-ray diffractograms obtained on multilayers B, C, D are presented in Fig. 3. The already mentioned similitude of the results for samples C and D still remains in XRD experiments. From the diffractograms, it is first inferred that crystalline Fe-based layers exhibit a [110] texture. Both (200) and (211) bcc reflections are absent, as well as satellite peaks around (110) line due to stacking periodicity. The absence of the latter peaks is mainly due to interdiffusion phenomena which took place during the annealing process.

Indeed the shift of the (110) peaks with respect to that for standard Fe (Fig.3) can be attributed to alloying effects. The peak width at half maximum is also determined as 1.9° for sample B and 3° for samples C and D. Line broadening is generally attributed to both coherent diffracting domain size and microstrains due to stacking faults, dislocations and point defects. Though a detailed deconvolution was not performed, we determined the average crystallite size using Scherrer equation and found 7.0 ± 0.7 and 4.5 ± 0.5 nm for samples B and C, D respectively. These values are greater than the Fe interlayer thicknesses but lower than the modulation periods of the as-prepared samples. The results are in agreement with those obtained from Mössbauer experiments which revealed the growth of broad interdiffusion layers. Moreover the XRD study proves the nanocrystalline character of the interdiffusion zone assumed from CEMS results; the assumption of an amorphous structure would be then ruled out. Finally the rocking curves, not shown here, indicate an equivalent misorientation of textured grains, with respect to the surface, of about 5° for the three samples. As a conclusion, both CEMS and XRD techniques proved to be good tools for studying interface effects in Fe/CoNbZr multilayers qualitatively and quantitatively. CEMS experiments performed at low temperature on the annealed samples would be also interesting to confirm the superparamagnetic behaviour of the interdiffusion zone.

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