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FERROMAGNETIC RESONANCE IN Fe/Zr MULTILAYERS

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We report our ferromagnetic resonance measurements for three series of Fe/Zr multilayers with thickness ratio of Fe to Zr sublayers equal to 2:1, 1:1, and 1:2, respectively. These measurements may serve as an example of employing ferromagnetic resonance technique as a sensitive probe for monitoring the magnetic phases formed at interfaces in the earliest stages of the interdiffusion reaction in multilayer Fe/Zr films for which the onset of the amorphization takes place during deposition due to thermodynamics of the system. The resulting structure may be regarded as strongly inhomogeneous one that includes distinct ferromagnetic phases related to the Fe atoms in various surroundings.
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1. Introduction

Formation of amorphous phase at interfaces of Fe/Zr multilayers (Mls) during a deposition process has been clearly demonstrated using the conversion electron Moessbauer spectroscopy (CEMS) [1–3] as well as using standard magnetic measurements [4]. It has been shown that Fe/Zr Mls can be produced in either predominantly crystalline or completely amorphous state depending on thickness of each constituent. In particular, for $n(\mathrm{Fe}) = n(\mathrm{Zr}) < 10$ monolayer, both sputter-deposited and electron-beam deposited Fe/Zr Mls were found to be entirely amorphous [2, 4]. Using CEMS [2], that serves as a local probe for determining the state of Fe, the non-magnetic (at room temperature) amorphous phase of FeZr, the bulk α-Fe, and the interfacial bcc α-Fe were distinguished. Kiauka et al. [3] showed that a large fraction of Fe atoms in a solid-state-reacted (SSR) amorphous phase are in a magnetically ordered state at low temperature.

According to our investigations, in the as-prepared Fe/Zr MLs consisting of Fe sublayers of the thickness ranged from 8 to 3 nm a complicated magnetic multiphase system exists as a result of the SSR taking place during the deposition. The aim of this contribution is to present new magnetic data for Fe/Zr MLs that can provide information on the structure of interfaces and the possible occurrence of various magnetic phases. Ferromagnetic resonance (FMR) technique was mainly applied for this purpose since its ability to probe various magnetic inhomogeneities has been well established.

2. Experimental details

Films were prepared by an alternate deposition sputtering technique using two Fe and Zr magnetically enhanced sources in a "face-to-face" configuration. The variable parameters characterizing the films were the modulation wavelength $1.2 \leq \lambda \leq 12$ nm ($\lambda = d_{Fe} + d_{Zr}$, where d_{Fe} and d_{Zr} are Fe and Zr sublayer thicknesses, respectively), and the sublayer thickness ratio $d_{Fe}/d_{Zr} = 0.5$, 1, and 2. The total thickness of the multilayers was kept constant at about 230 nm and was measured independently with the X-ray fluorescence analysis [5] and Tolansky interference method.

Both small and large angle X-ray diffraction in the Bragg–Brentano geometry were used to characterize the composition modulation and crystallinity, respectively. Small-angle X-ray diffraction showed several superstructure peaks indicating a good periodicity. Nevertheless, a number of peaks decreases with the sublayer thickness implying intermixing at interfaces. Since an amorphization takes place during deposition, a real thickness of both the Fe and Zr sublayers is lower than their nominal thickness and an interface amorphous alloy occupies a significant volume of the films. Therefore, both d_{Fe} and d_{Zr} should be rather regarded as nominal thicknesses which reflect the "amount" of both constituents deposited throughout the alternate deposition sequences.

We have investigated three sets of samples with $d_{Fe}/d_{Zr} = 0.5$, 1, and 2 which yield the mean composition of $Fe_{50}Zr_{50}$, $Fe_{67}Zr_{33}$, and $Fe_{80}Zr_{20}$, respectively. Magnetic measurements were performed using FMR, and vibrating sample magnetometer (VSM). Field derivative of FMR absorption was recorded in the temperature range from about 120 K to 300 K using a standard EPR spectrometer operating at 9 GHz. Standard resonance conditions [6] for magnetic field applied perpendicularly or parallel to the film surface were used for evaluation of the effective magnetization $4\pi M_{eff} = 4\pi M_s - H_U$ and the g-factor for each resonance of the spectra. The effective magnetization includes the demagnetizing field $4\pi M_s$ as well as the uniaxial anisotropy term H_U that describes the surface and volume anisotropy fields. The intensity of each resonance in FMR spectrum (doubly integrated field derivative of the absorption) provides us with valuable information as well since it is proportional to the ($4\pi M_s \times$ volume) product for each constituent present in a film [7]. The magnetic moment was measured at room temperature.

3. Results and discussion

The results of the static magnetic measurements with VSM for the three sets of films are shown in Fig. 1, where the relative magnetic moment $m(d_{Fe})/m(\infty)$ versus the Fe sublayer thickness d_{Fe} is plotted. Here, $m(d_{Fe})$ is the magnetic moment per unit area, and $m(\infty)$ is the magnetic moment per unit area of our standard continuous Fe film 156 nm thick. Generally, all data fall into a one universal curve. A drastic decrease in $m(d_{Fe})/m(\infty)$ is seen for $3 < d_{Fe} < 8$ nm and is regarded to be caused by an amorphous alloy formation at the interfaces between Fe an Zr by SSR during the deposition process. This large decrease in $m(d_{Fe})/m(\infty)$ is presumably additionally intensified via a significant thickness dependence of M_s that has been frequently observed in thin films [8].

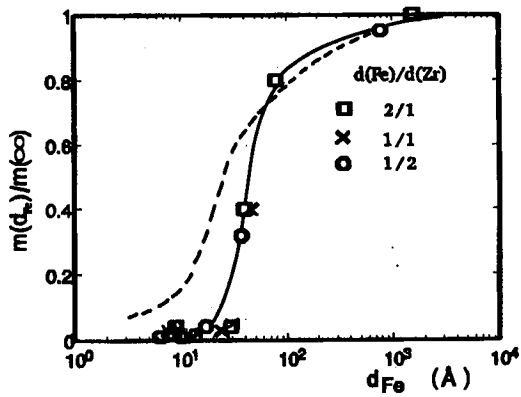

Fig. 1. Relative magnetic moment $m(d_{\mathrm{Fe}})/m(\infty)$ for three series of Fe/Zr MLs as a function of the nominal thickness d_{Fe}. The dashed curve depicts the dependence of the relative magnetization $4\pi M_{\mathrm{eff}}(d_{\mathrm{Fe}})/4\pi M_{\mathrm{eff}}(\infty)$ on d_{Fe}. $4\pi M_{\mathrm{eff}}(\infty) = 22$ kG of a thick Fe film.

In conclusion, it follows from our static magnetic measurements that, at room temperature, our sputter-deposited Fe/Zr MLs with the nominal thickness $d_{\mathrm{Fe}} < 2$ nm are paramagnetic and amorphous.

A more local insight into the nature of all magnetic phases existing in the available temperature range could be achieved from an analysis of FMR spectra. Figure 2 shows typical FMR spectra of Fe/Zr MLs with $d_{\mathrm{Fe}}/d_{\mathrm{Zr}} = 1/1$ taken at room temperature for the parallel configuration. With decreasing d_{Fe}, the FMR spectra transform from single-resonance spectra characteristic of thin Fe films Fe(2.9 nm)/Zr(2.9 nm) to strongly asymmetric or multi-resonant ones, characteristic of inhomogeneous or multiphase magnetic films. The asymmetric lineshape of the spectrum of Fe(2.4 nm)/Zr(2.4 nm) MLs can be associated with a significant compositional gradient within magnetic phases: from α-Fe to a magnetic phase with $4\pi M_{\mathrm{eff}} = 0$. The intensity and the resonance field position of the low field resonance of the spectrum for Fe(1.3 nm)/Zr(1.3 nm) merely depends on temperature. We attribute this resonance to α-Fe at interfaces. A rough calculations based on the comparison of the intensities of resonances observed in the FMR spectra at low temperatures led us to the conclusion that the amorphous phase is dominant; the volume of α-Fe amounts only to few percent of the volume of the amorphous phase. Thus, taking into account such a small fraction of α-Fe that exists in the Fe/Zr films with the nominal thickness $d_{\mathrm{Fe}} < 2$ nm, we may argue that it rather consists of small Fe particles than continuous Fe sublayers. Therefore, it seems that the interfaces in these films are rather rough. It would also explain the weak but visible dependence of $4\pi M_{\mathrm{eff}}$ dependence on temperature for α-Fe. Therefore, we may argue that in the region of an abrupt decrease in $m(d_{\mathrm{Fe}})/m(\infty)$ the Fe/Zr MLs, must be looked on as a multicomponent system consisting of the crystalline Fe and Zr and an amorphous FeZr alloy phase. This conclusion agrees with the CEMS measurements [1–3].

For such multiconstituent system, from the resonance positions taken at the perpendicular and parallel configuration, the values of $4\pi M_{\rm eff}$ and the g-factor were determined for each constituent which shows up in the spectrum as a clearly distinct resonance. When the maximal magnetic field available (11 kOe) was not sufficient for FMR measurements at the perpendicular configuration, the g-factor was chosen to be equal 2.08 of pure Fe. The dependence of $4\pi M_{\rm eff}(d_{\rm Fe})/4\pi M_{\rm eff}(\infty)$ versus $d_{\rm Fe}$ related to the Fe phase is depicted in Fig. 1 by a dashed curve with no experimental points, for clarity. $4\pi M_{\rm eff}(\infty)$ was assumed to be equal to 22 kG for a standard Fe film 156 nm thick.

For the thickness range where, according to the static measurements $m(d_{\rm Fe})/m(\infty) = 0$, our Fe/Zr MLs with $d_{\rm Fe}/d_{\rm Zr} = 1:1$ seemed to be full amorphous and paramagnetic at room temperature. Nevertheless, we observed some FMR resonances related to the remaining ferromagnetic phases. Typical temperature dependencies of $4\pi M_{\rm eff}$ for two constituents characterizing the film with $d_{\rm Fe}/d_{\rm Zr} = 1.3/1.3$ nm is displayed in the inset of Fig. 2 as an illustration of the magnetic behaviour of Fe/Zr MLs with $d_{\rm Fe} < 2$ nm. The upper curve shows only relatively weak dependence on temperature and, therefore, the resonance line placed at low field range (the dashed curve in Fig. 2) can be attributed to an unreacted α-Fe phase. The bottom curve in inset of Fig. 2 is characterized by the

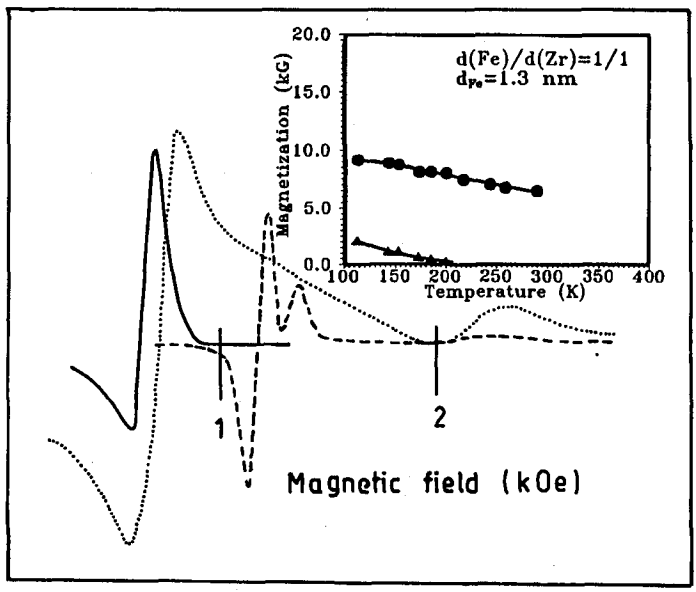

Fig. 2. Typical room temperature FMR spectra of Fe/Zr MLs with $d_{\rm Fe}/d_{\rm Zr} = 1/1$. Continuous, dashed, and dotted curves display FMR spectra taken at the parallel configuration for films with $d_{\rm Fe} = 2.9$, 2.4, and 1.3 nm, respectively. The inset shows the temperature dependence of the effective magnetization $4\pi M_{\rm eff}$ of two constituents which are present in the Fe/Zr film with $d_{\rm Fe} = d_{\rm Zr} = 1.3$ nm.

Curie temperature of about 200 K and, thus, may be attributed to the $Fe_{67}Zr_{33}$ amorphous alloy phase [9].

4. Conclusions

The as-deposited Fe/Zr MLs should be looked on as a multicomponent system consisting of the crystalline Fe and Zr sublayers, the Fe fraction at interfaces, and the amorphous FeZr alloy of composition depending on both the Fe layer thickness d_{Fe} and the thickness ratio d_{Fe}/d_{Zr}. A variety of magnetic constituents related to Fe atoms in different surroundings has been revealed using FMR technique. These results agree with the CEMS investigations which revealed the Moessbauer spectra attributed to the interface region between α-Fe and the amorphous FeZr [1, 2] and showed that two different Fe states exist in the amorphous phase due to a Fe concentration gradient across the amorphous phase [3]. From the analysis of both the resonance positions and their intensities, we have shown that the remaining unreacted α-Fe in the Fe/Zr MLs rather consists of small grains than continuous sublayers.

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