Combined Analysis of Static and Dynamic Magnetic Characteristics of Multilayer CoFeZr/α-Si Nanostructures

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Abstract—The combined method of static and dynamic magnetic measurements is used to study nano-dimensional multilayer amorphous CoFeZr/ α -Si films. It is established that at the thickness of magnetic layers x = 5–12 nm their magnetization does not differ from that of the bulk material. It is shown that as *x* is lowered to 2–3 nm, the magnetization of magnetic layers decreases, which may be due to the formation of mixed layers containing nonmagnetic silicides. At a thickness of nonmagnetic interlayers of less than 1 nm the features characteristic of a weak antiferromagnetic interaction of neighboring layers are observed.

Key words: nanostructure, multilayers, discontinuous multilayers.

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Recently magnetic nanostructures containing contacting layers of ferromagnetic metal and semiconductor have attracted the attention of researchers as a promising material for spintronics devices, e.g., when creating spin-field transistors [1, 2]. One of the problems arising when manufacturing multilayer structures with Si interlayers is the formation of silicides on the metal– semiconductor interface [3].

We studied multilayer CoFeZr/ α -Si structures prepared by ion-beam sputtering of targets of the amorphous Co₄₅Fe₄₅Zr₁₀ alloy and amorphous α -Si in an argon atmosphere (with a purity of 99.992%) with the addition of hydrogen at the 2 kV voltage on the glassceramic and silicon substrates at temperature not exceeding 50°C (see [3]). The thickness *x* of magnetic layers specified at the deposition varied from 1 to 12 nm and the thickness *y* of the nonmagnetic silicon interlayers from 0.5 to 5 nm. The total number of bilayers *z* = 40.

Three series of samples with the thickness x = 1.9– 2.3, 3 nm (thin, series 1 and 2) and 5–12 nm (thick, series 3) and variable interlayer thickness y (0.5–5 nm) were considered. For all samples the ferromagnetic resonance (FMR) spectra depending on the orientation of the external field **H** with respect to the film plane and magnetization curves were measured. FMR spectra were measured on an EPR spectrometer at a frequency of f = 9.27 GHz. Static measurements were carried out on a vibration magnetometer in fields up to 10 kOe. Hysteresis loops were measured in the layer plane in two mutually perpendicular directions. The mean saturation magnetization of the multilayer structure $I_m = M[Sz(x + y)]$ and magnetization of magnetic layers $I_l = M/(xzS)$ were determined from the magnetic moment of the unit sample area (*M/S*) in the saturation region and data on its total thickness z(x + y).

According to the static data, in samples with x > 1 nm the interaction between layers is ferromagnetic. The form and quantitative parameters of the loops in the series considerably depend on the values of x and y specified during preparation and on the ratio x/(x + y). Figure 1 shows, as an example, the change of the form of the hysteresis loops of the structures with the



Fig. 1. Evolution of the form of the hysteresis loops of $Co_{45}Fe_{45}Zr_{10}/\alpha$ -Si nanostructures at a, y = 0.97 nm; b, y = 1.7 nm; c, y = 4.45 nm. (Loops are measured in two mutually perpendicular directions).



Fig. 2. Dependences of H_{\parallel} on *y*. Curve *1*, series 1; curve *2*, series 2; curve *3*, series 3.

increase in the y value. A gradual transition from the complicated loop form with an indistinct step and coercive force $H_c \approx 6-7$ Oe to the form typical for soft magnetic films with the uniaxial anisotropy in the film plane and $H_c < 1$ Oe is observed. The mean magnetization I_m gradually decreases. However, the experimental I_m values noticeably differ from those predicted by the calculations under the assumption that the real thickness of the magnetic layers is that specified at the synthesis and the saturation magnetization is the magnetization of the bulk $Co_{45}Fe_{45}Zr_{10}$ alloy (estimated from the FMR data as 1050–1140 G).

The discrepancy between the calculated and experimental I_m values may evidence that during the preparation of the multilayer nanostructure new phases are formed (e.g., various types of silicides). To clarify the structural features of these samples the results of the static and dynamic measurements of magnetic characteristics were analyzed in detail.

Figure 2 shows the dependences of the resonance fields of the homogeneous-type precession (H_{\parallel}) on the thickness of the nonmagnetic interlayer for the parallel orientation of **H** with respect to the film plane in all series under investigation. The dashed lines in Fig. 2 show data for the samples with y < 1 nm (the relevant results will be discussed below). The plots show that at large *x* (of about 5–12 nm), i.e., at the formation of continuous layers, H_{\parallel} values practically do not depend on the thickness of the nonmagnetic inter-layer and are 780–800 Oe. The static measurements showed that the anisotropy in these samples is small. By using the Kittel formula

$$\left(\omega/\gamma\right)^2 = H_{\parallel}(H_{\parallel} + 4\pi I_0), \tag{1}$$

 $(\omega = 2\pi f \text{ and } \gamma \text{ is the giromagnetic ratio, in our case of the order of 2.7–2.9 MHz/Oe) we obtain the magnetization <math>I_0 = 1050-1140 \text{ G}$. The same I_0 values calculated from H_{\parallel} were obtained for the bulk samples (I_0) and thin-film samples sputtered without the amorphous sil-



Fig. 3. Dependences of the resonance fields of different lines in the magnetostatic vibration spectrum on the interlayer thickness (y). •, \mathbf{H}_{\perp} ; \Box , H_{\parallel} . Solid lines connect resonances of supposedly similar vibration types. Bold lines show the homogeneous precession. a, series 1; b, series 2.

icon interlayers. These values are close to the magnetization of the magnetic layers I_l calculated from the magnetization curves (920–1280 G) of the samples in this series.

As the *x* value is decreased, H_{\parallel} starts to depend on the thickness of the nonmagnetic layers: the smaller *x* is, the more noticeable the dependence is. The minimal H_{\parallel} values for x = 2-3 nm are observed for $y \approx 1$ nm, when the film composite of a ferromagnetic material with amorphous Si inclusions is formed.

The static measurements showed that the magnetization of the magnetic layer in the series with $x \approx 2$ and 3 nm is lower than that in the bulk samples.

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No.	x, nm	y, nm	<i>M/S</i> *, G/nm	I _m , G	<i>I</i> _{<i>l</i>} , G	<i>I</i> ₀ , G	<i>x</i> ', nm
Series 1							
1	1.9	0.8	52345	485	690	567	1.24
2	2.12	1.2	54020	410	640	508	1.28
3	2.25	1.66	46818	300	520	414	1.11
4	2.2	1.9	50094	290	540	335	1.19
5	2.2	2.2	36146	205	410	272	0.86
6	2.29	2.45	32342	170	350	221	0.77
7	2.3	2.7	23250	115	250	169	0.55
8	1.94	1.7				506	
9	1.94	1.76				490	
10	1.95	1.83				470	
11	1.95	1.9				450	
12	1.95	2.0				444	
13	1.96	2.1				416	
Series 2							
1	2.71	1.7	57111	320	530	622	1.38
2	2.87	2.54	56335	260	490	508	1.36
3	2.93	3.1	48069	200	410	446	1.16
4	2.97	3.57	47785	180	400	399	1.15
5	2.99	4.14	39137	140	330	349	0.95
6	2.93	4.45	36794	120	310	370	0.89

Table

* Error in measuring *M/S* was about 10–15%.

Figure 3a and b presents the dependences of the resonance fields for different types of spectral vibrations on the thickness of the nonmagnetic interlayer excited in the parallel (H_{\parallel}) and perpendicular (H_{\perp}) orientations of the **H** field with respect to the film plane in the samples with $x \approx 2$ and 3 nm. The complexity of the system due to the internal inhomogeneities does not allow us to completely interpret the spectrum. However, the comparison of Fig. 3a and b shows that the internal fields of the samples in these series differ. At the thickness $x \approx 2$ nm for the fixed interlayer thickness the spectral lines are observed in a wide field range. For some vibration types the resonance fields for the two **H** directions are close to each other. X-ray data [4] showed that for these x values there are no distinct boundaries between the layers in the samples and magnetic layers have an island structure. Direct contacts between magnetic layers are possible. This should result in large inhomogeneities of the internal magnetic field. At $x \approx 3$ nm the formation of the layers is mainly finished, the structure transfers from an island one to a continuous one and the internal fields become more homogeneous. Their magnetization at y > 3 nm is much higher than in the series with $x \approx 2$ nm; therefore, for such samples the resonance field values at the circumferential and normal magnetizations strongly differ.

At present there are two approaches to the description of the FMR phenomenon in the multilayer structures. In the first approach, the behavior of the magnetization vector is considered separately in each magnetic layer. It may be described by the motion equation of the layer magnetic moment $dI/dt = -\gamma I \times H_{\text{eff}}$, where *t* is time. The influence of the remaining part of the film is taken into account by introducing the effective magnetic fields H_{eff} determined via the magnetic energy of the whole system [5].

In the second approach, the multilayer structure is considered as an effective medium with the mean magnetization $I_{\rm m} = I_l x/(x + y)$.

The table presents the values of the mean saturation magnetization I_m of the sample, magnetization of the magnetic layer I_l and magnetization I_0 calculated from the FMR data (Eq. (1)) for series 1 and 2. One can see from the Table that the calculation data obtained from the static and microwave measurements coincide well only under the assumption that the behavior of the magnetization vector is determined by the magnetization of the magnetic layer, which is lower than that in the bulk samples.

The observed decrease of the magnetization has a twofold explanation. First, the electron systems of the silicon interlayer and the magnetic layer may interact in



Fig. 4. Dependences of the magnetization on *y* in samples of series $1. \times, I_{\text{weak}}; \bullet, I_{\text{m}}; \bigcirc, I$ from the FMR data; solid line, calculated according to Eq. (2).

such a manner that the electron system of the magnetic layer changes. For example, in [6] it was shown that the change of the magnetic moment of the cobalt ions depends on the structure of the interface. In particular, in Co/Rh samples the magnetic moment of the cobalt ions does not change, even at strong intermixing of the Co and Rh ions in the interface. At the same time, it strongly decreases in the interface with the increase in the Ru concentration. The same result was obtained in [7] on the Fe/V samples in which, at an iron layer thickness of 2 Å, the exchange interaction and the local magnetic moments of Fe ions were absent. However, in our case such a contribution of the thin interface layers (<0.2 nm) to the total magnetic moment of the layer 2– 3 nm thick would be too small to be registered in the static measurements.

On the other hand, as mentioned above, the formation of silicides may cause the apparent decrease of the magnetization of the magnetic layers. In [8] it was shown that during the ion-plasma sputtering of the multilayer $Fe_{89}Co_{11}$ -Si structures at the deposition of the $Fe_{89}Co_{11}$ on the silicon interlayer an intermediate layer 1.5 nm thick with the composition close to the metal monosilicide ($Fe_{89}Co_{11}$)_{0.5}Si_{0.5}) is formed. At the deposition of the Si interlayer on the $Fe_{89}Co_{11}$ a $Fe_{89}Co_{11}$)_{0.67}Si_{0.33} layer 1.9 nm thick is formed.

We carried out calculations of two types. In the first variant, the thickness of the magnetic layer necessary to provide the experimental values of the magnetic moment of the sample under the condition that the magnetization of the layer is the magnetization of the bulk material I_{0M} was estimated. The thickness was calculated according to the formula x' = M/(zSI), where x' is the thickness of the magnetic layer with the magnetization I_{0M} . The table shows that the change of x should be



Fig. 5. Dependences of the magnetization on *y* in samples of series 2. Notations are the same as in Fig. 4.

considerable. Moreover, in this case it is necessary to assume that the magnetic layer should be considered as an effective medium (not taking into account the nonmagnetic interlayer) composed of two layers one of which has the magnetization of the bulk material and the other has zero magnetization. Otherwise, the results of the spectral measurements of the resonance fields will differ strongly from the calculated values.

In the second variant it was supposed that a part of the atoms of the metal layer form nonmagnetic silicides. The part of the remaining magnetic atoms is determined by

$$1 - N_{\rm Si}/N_{\rm Me} = 1 - y\rho_{\rm Si}\mu_{\rm Me}/(x\rho_{\rm Me}\mu_{\rm Si}), \qquad (2)$$

where ρ and μ are the density and molar mass of the corresponding materials, respectively. We used the following (for the metal, mean) values: $\rho_{Si} = 2.1$ g/cm³, $\rho_{Me} = 8.1$ g/cm⁻¹, $\mu_{Si} = 28$ g/mol and $\mu_{Me} = 58$ g/mol. The magnetization of the magnetic layer without silicides is 1050 G. The magnetization with silicides is calculated under the assumption that it is proportional to the number of the remaining metal atoms. Naturally, this model fits samples with thinner and more porous magnetic layers better. In spite of the roughness of the model, the calculated and experimental values coincide satisfactorily (Figs. 4 and 5).

The results obtained for the samples with the same composition of the magnetic layers and the dielectric SiO₂ interlayer favor this model. In this case, silicon is bound by oxygen and the conditions for the formation of silicides are less favorable. this results in the samples with the SiO₂ inter-layer having less silicides than those with the same *x* and *y* values but the semiconductor α -Si interlayer. The magnetization of the latter samples is higher and, as a result, they have lower H_{\parallel} values. For example, in the sample with the α -Si interlayer x =

2.97 nm, y = 4.3 nm, $H_{\parallel} = 1530$ Oe, and in the sample with the SiO₂ interlayer, x = 3.1 nm, y = 4.5 nm, and $H_{\parallel} = 860$ Oe. The magnetization of the magnetic layers of the samples with the SiO₂ interlayer calculated from the magnetization curves was about 1100–1200 G (for the samples with the α -Si interlayer, I = 350 G) somewhat higher than the values obtained from Eq. (1), taking into account the experimental H_{\parallel} values (I = 950 G).

Clearly, quantum effects play a noticeable role in the region y < 1 nm. The static measurements showed that as the interlayer thickness is decreased, the magnetization of the samples decreases and the H_{\parallel} value increases. This may be partially due to the island structure and the large number of paramagnetic or superparamagnetic particles. On the other hand, the influence of nano-dimensional effects is possible. The form of the magnetization curves (Fig. 1) show that in these samples phases with different coercive forces and a weak antiferromagnetic interaction between the layers are present. Thus, for example, the saturation magnetization field of the samples with x = 2.5 nm and y = 0.8 nm exceeds 3kOe. At these inter-layer thicknesses a negative magnetic resistivity is observed reaching its maximum at y = 0.7 nm [3]. According to the magneto-optical data [9], at y = 0.5 nm the curves of the equatorial Kerr effect are similar to those observed in composites at magnetic phase concentrations slightly below the percolation threshold.

In summary, let us emphasize the main results of the present work. The static and high-frequency properties of the multilayer nano-dimensional $Co_{45}Fe_{45}Zr_{10}/\alpha$ -Si structures were studied. The calculations and experimental FMR data agree well under the assumption that the behavior of the magnetization vector is determined by the magnetization of the magnetic layer.

The formation of the silicides plays an important role in the formation of the properties of the multilayer structures. A calculation accounting for the influence of silicides on the characteristics of the systems under investigation is proposed. The calculated and experimental data agree well. The results obtained for the samples with the same composition but with the dielectric SiO_2 interlayer favor this model.

The interaction of the layers is mainly ferromagnetic. However, as the nonmagnetic interlayer y < 1 nm decreases, the transition to the weak antiferromagnetic interaction is observed.

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