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ELECTRICAL AND MAGNETIC PROPERTIES

Micromagnetic Structure of Soft Magnetic Nanocrystalline Fe-Based Films

E. V. Harin and E. N. Sheftel

Baikov Institute of Metallurgy and Materials Science, Russian Academy of Sciences, Leninskii pr. 49, Moscow, 119991 Russia e-mail: sheftel@imet.ac.ru, harin-eugene@ya.ru Received July 7, 2014; in final form, February 10, 2015

Abstract—Results of a quantitative determination of parameters of the micromagnetic structure of nanocrystalline Fe, $Fe_{95}Zr_5$, $Fe_{90}N_{10}$, and $Fe_{85}Zr_5N_{10}$ films prepared by magnetron sputtering have been reported. The magnetocrystalline (K_1), magnetoelastic (K_{ME}), magnetostatic (K_{MS}), and surface ($K_{a,s}$) anisotropy constants have been shown to be components of the effective local anisotropy (K_{eff}) constant determined experimentally. The shape of hysteresis loops is determined by the existence of two main components of macroscopic effective magnetic anisotropy, one of which is caused by local (within a nanograin) magnetic anisotropy averaged over the exchange interaction length, while the other is related to magnetoelastic anisotropy due to residual macrostresses.

Keywords: nanocrystalline, soft magnetic FeZrN films, magnetic anisotropy, magnetic structure **DOI**: 10.1134/S0031918X15080074

INTRODUCTION

Films of soft magnetic Fe–Zr–N alloys (with the high iron content equal to \sim 80–95 at %), which have nanocrystalline structure, assure a unique combination of magnetic and mechanical properties [1], which is required for their application in miniature devices of advanced microelectronics.

Depending on the zirconium and nitrogen contents in the films, their phase composition can comprise the main ferromagnetic α -Fe-based phase, ferromagnetic iron nitrides, Fe-Zr intermetallics, and one or several nonferromagnetic phases, the formation of which is possible for the ternary Fe–Zr–N system. It is known [2] that the magnetization process in ferromagnets is determined by the effective magnetic anisotropy. In the case of uniaxial anisotropy, it is characterized by the $K_{\rm eff}$ anisotropy constant; in the total case, the anisotropy includes the magnetocrystalline (the K_1 constant), magnetoelastic (the K_{ME} constant), surface (the K_s constant), and magnetostatic (the K_{MS} constant) components. The possibility of quantitativly estimating the components assures conditions for reasonably controlling the level of magnetic properties. Generally, in the literature, the effect of grain size (magnetocrystalline energy) on the magnetic properties of nanocrystalline ferromagnets is considered [3]. There are a few works devoted to the cooperative effect of magnetocrystalline and magnetoelastic energies in the literature [3]. The surface anisotropy is usually considered individually [4]. Studies related to the role of magnetostatic energy,

which were performed in 30th and 40th of the last century, deal with bulk materials [5]. No data related to the investigation and quantitative estimation of all the aforementioned contributions to the K_{eff} constant of Fe–Zr–N films are available.

The aim of the present study is to quantitatively estimate specific energies of the magnetocrystalline, magnetoelastic, surface, and magnetostatic anisotropy and their effect on the coercive force (H_c) of Fe and Fe-based (alloyed with N and Zr) films.

THEORETICAL BASES

The magnetocrystalline energy is determined by crystallographic anisotropy of a material and is a constant (K_1) for an associated chemical composition of phase (substance, solid solution, alloy, compound, etc.) and temperature. The magnetoelastic energy $(K_{\rm MF})$ is determined by the saturation magnetostriction (λ_s) of a material and mechanical stresses (σ) that exist in the material, i.e., $K_{\text{ME}} = (3/2)\lambda_s \sigma$. The surface anisotropy (K_s) is caused by the violation of atomic arrangement symmetry at the surface of ferromagnetic grains. The magnetostatic energy $(K_{\rm MS})$ is caused by magnetic poles formed at surfaces of nonmagnetic grains within the magnetic matrix. Each of the energies corresponds to its own anisotropy field, the general form of which is $H_K = 2K/M_s$, where M_s is the saturation magnetization of ferromagnetic phase. The $H_{\rm c}$ coercive force of ferromagnetic materials, which is related to the

Chemical compo- sition, at %	Fe	Fe ₉₀ N ₁₀	Fe ₉₅ Zr ₅	$Fe_{85}Zr_5N_{10}$
Film thickness, nm	370	300	370	500

 Table 1. Chemical composition and thicknesses of studied films

anisotropy field $H_c \approx H_K/2$, is usually used to characterize the magnetic properties of ferromagnets.

The feature of nanocrystalline ferromagnets (with a grain size of less than ~50 nm) consists in the fact that their grain size is less than the length of the exchange interaction [3]. This causes the relatively uniform parallel alignment of magnetization over distances higher than the grain size, which forms so-called stochastic domains [6]. Within stochastic domains, the effective local anisotropy ($K_{\rm eff}$) of randomly oriented grains is partially suppressed by the exchange interaction. The grain size of these ferromagnets determines the autocorrelation radius R_c , and the exchange-interaction length determines the stochastic-domain radius R_L . Local (within the grain) components of $K_{\rm eff}$ correspond to radius R_{c} , and the components of macroscopic anisotropy $(\langle K_{eff} \rangle)$ correspond to radius R_L . The effect of local energy on the properties of area of radius R_L is given analytically by equation $\langle K_{\text{eff}} \rangle = K_{\text{eff}}$ $(R_c/R_L)^{3/2}$ [6].

It should be noted that the magnetic structure of these ferromagnets can have several characteristic radii R_L (characteristic lengths of magnetic orientation coherency), in particular the exchange-interaction length for an ensemble of grains, wavelength of macrostresses, etc. In the case of comparable volumes occupied by magnetic elements with different characteristic radii R_L (and, therefore, with different density of magnetic-anisotropy energy), the coercive force of a nanocrystalline ferromagnet is determined by the anisotropy field H_K of an element of magnetic structure (R_L) having the maximum size and, therefore, the minimum magnetic anisotropy energy.

EXPERIMENTAL

The films to be studied were deposited on cover glass substrates by magnetron sputtering of cast Fe and Fe-5 at % Zr targets. The residual gas pressure in the sputtering chamber was $\sim 2 \times 10^{-6}$ Torr. The argon pressure is $\sim 3.5 \times 10^{-3}$ Torr. The nitrogen pressure during preparation of nitrogen-containing films was $\sim 2.5 \times 10^{-4}$ Torr. The film thicknesses were determined using an interference microscope. The chemical composition of films was determined using an FEI Quanta 200 scanning electron microscope equipped with an attachment for energy dispersion X-ray microanalysis (EDAX). Table 1 shows the chemical composition and thickness of the films. The structure of films was studied by X-ray diffraction analysis using a DRON-3 diffractometer and Cu $K\alpha$ radiation. X-ray diffraction patterns were processed using special software [7]. The grain size $2R_c$ and true microstrains ε of phases present in the films were estimated using the physical broadening of X-ray diffraction reflections. Macrostresses (σ) in the films were calculated using lattice parameters from two reflections from planes differing in elastic properties [8]. Static magnetic properties were measured in fields up to 20 kOe using a vibrating-sample magnetometer. Parameters of magnetic structure (effective local-anisotropy field H_a and exchange field H_R) were estimated by correlation magnetometry [6]. The field dependence of magnetostriction of films was measured in fields up to 80 Oe using an atomic force microscope and an original cantilever technique [9]. All measurements were performed at room temperature.

RESULTS AND DISCUSSION

Phase Composition, Structure, and Macrostresses

According to X-ray diffraction data, all studied films are nanocrystalline. Their phase composition includes the ferromagnetic α -Fe-based phase and a small amount of nonferromagnetic Fe₂N nitride that is presented additionally in nitrogen-containing films (Table 2).

The X-ray diffraction patterns of all films contain reflections from the (110) and (200) planes. This

Film	Phase composition, vol %	Lattice parameter, Å	Grain size $2R_c$, nm	Microstrain ε, %
Fe	bcc α-Fe	2.824 ± 0.003	39 ± 11	0.56 ± 0.04
Fe ₉₀ N ₁₀	bcc α -Fe(N) ~ 95	2.873	15	0.53
	$Fe_2N \sim 5$	a = 2.759; c = 4.429	78	0.07
Fe ₉₅ Zr ₅	bcc α -Fe(Zr)	$a = 2.857 \pm 0.008$	34 ± 3	0.23 ± 0.02
$\mathrm{Fe}_{85}\mathrm{Zr}_5\mathrm{N}_{10}$	bcc α -Fe(N,Zr) ~ 93	$a = 2.899 \pm 0.007$	11 ± 1	0.24 ± 0.09
	$Fe_2N \sim 7$	a = 2.738; c = 4.560	35	1.05

Table 2. Phase composition and structural characteristics of the studied films

allows us to estimate macrostresses (σ) in the films by the equation

$$\sigma_{\rm res} = \frac{a_{h_1 k_1 l_1} - a_{h_2 k_2 l_2}}{2(a_{h_2 k_2 l_2} K_{h_1 k_1 l_1} - a_{h_1 k_1 l_1} K_{h_2 k_2 l_2})}.$$
 (1)

where $a_{h_1k_1l_1}$ and $a_{h_2k_2l_2}$ are the lattice parameters of the α -Fe phase, which were determined using the (110) and (200) reflections, respectively; $K_{h_1k_1l_1}$ and $K_{h_2k_2l_2}$ are the elastic constants for the (110) and (200) crystallographic planes; $K_{hkl} = (\nu/E)_{hkl}$; and ν and Eare the Poisson's ratio and Young modulus for the associated crystallographic planes (we take the magnitudes equal to those for pure iron) [8]. Table 3 shows the used literature data on the $K_{h_1k_1l_1}$ and $K_{h_2k_2l_2}$ constants [8] and macrostresses σ in the film plane, which were calculated by Eq. (1).

Static Magnetic Properties and Parameters of Magnetic Structure

To determine the saturation magnetization (M_s) of each samples, the experimental magnetization curve M(H) measured in fields of more than 2 kOe was plotted on coordinates $M-H^{-2}$ (Fig. 1a). The obtained linear portion of the curve was described by the Akulov's law as follows [5]:

$$M = M_{\rm s} \left[1 - \frac{1}{2} \left(\frac{D^{1/2} H_{\rm a}}{H} \right)^2 \right].$$
 (2)

where $D^{1/2}H_a$ is the root-mean-square fluctuation (within the R_c radius) of the effective local-anisotropy field and D is the dispersion of anisotropy axes, which is equal to 1/15 for randomly oriented uniaxialanisotropy axes. The uniaxial symmetry was taken based on the following two assumptions: (1) the existence of contributions of specific energy of the magne-

Table 3. Lattice parameters $(a_{110} \text{ and } a_{200})$ of the α -Febased phase, elastic constants K_{hkl} , and macrostresses (σ) for studied films

Film	Fe	Fe90N10	Fe ₉₅ Zr ₅	$\mathrm{Fe}_{85}\mathrm{Zr}_5\mathrm{N}_{10}$		
<i>a</i> ₁₁₀ , Å	2.8527	2.8679	2.8893	2.8986		
K_{110} , 10^{-4} GPa ⁻¹	-13.7					
<i>a</i> ₂₀₀ , Å	2.8401	2.8911	2.8712	2.9032		
K_{200} , 10^{-4} GPa ⁻¹	-28.7					
σ, MPa	1466	-2717	-2076	-530		

toelastic $K_{\rm ME}$, surface $K_{\rm s}$, and magnetostatic $K_{\rm MS}$ anisotropy to the effective local anisotropy, which on default have a uniaxial symmetry, and (2) these contributions and magnetocrystalline anisotropy are of the same order of magnitude. The factor of 1/2 (Holstein-Primakoff correction) in Eq. (2) is necessary if measurements are performed in fields $H \ll 4\pi M_s$ (that for iron is about 20 kOe). This correction is necessary due to the internal magnetic field, which is caused by the magnetization misorientation in grains [5]. The extrapolation of the linear dependence (Eq. (2)) plotted on coordinates $M-H^{-2}$ (Eq. (2)) to $H^{-2}=0$, i.e., to $H = \infty$ gives the saturation magnetization value. In the used magnetic fields (up to 10 kOe), the contributions from diamagnetic substrate and film magnetization paraprocess (Holstein-Primakoff paraprocess) to the magnetization curve are negligible and, therefore, were not taken into account.

Table 4 shows the saturation magnetization (M_s) , coercive force (H_c) , and relative remanence (M_R/M_s) for all films under study. Hereinafter, errors of measurements were determined using the spread of experimental points in hysteresis loops. It can be seen that the saturation magnetization of the Fe film is close to that of iron single crystal and equals 1707 G [10]. The alloying of Fe films with zirconium and nitrogen leads to a decrease in their saturation magnetization.



Fig. 1. (a) Magnetization as a function of H^{-2} and (b) field dependence of magnetization dispersion for the Fe₉₀N₁₀ film.

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Film	$M_{\rm s}, {\rm G}$	$H_{\rm a}$, Oe	H_R , Oe	$\langle H_{\rm a} \rangle$, Oe	$D^{1/2}\langle H_{\rm a}\rangle$, Oe	H_c , Oe	M_R/M_s	R_L/R_c
Fe	1683 ± 8	1200 ± 400	1700 ± 700	7.27 ± 0.89	1.88 ± 0.23	13 ± 2	0.75	29.74
Fe90N10	1332 ± 20	1800 ± 400	1500 ± 400	54 ± 5	14 ± 1.3	90 ± 5	0.75	10.67
Fe ₉₅ Zr ₅	1598 ± 9	800 ± 100	280 ± 40	320 ± 100	83 ± 26	45 ± 5	0.34	1.82
$\mathrm{Fe}_{85}\mathrm{Zr}_5\mathrm{N}_{10}$	1310 ± 7	1600 ± 100	800 ± 50	220 ± 36	57 ± 9	40 ± 5	0.37	3.64

Table 4. Magnetic properties and parameters of local (within grain) and macroscopic magnetic structures of studied films

According to [6], the field dependence of magnetization dispersion $d_{\rm m} = 1 - M/M_{\rm s}$ in the region of approach to the saturation contains information about effective parameters of the magnetic structure. In accordance with the theory of correlation magnetometry [6], for all studied samples, the dependence of magnetization dispersion $d_m(H)$ plotted on the log-log scale is described in high fields by two power functions intersecting at field H_R . Figure 1b shows an example of processing of the magnetization curve M(H) by the correlation magnetometry method. In fields $H > H_R$, exchange correlation between the magnetization vectors in neighboring grains is not exist; in this case, the magnetization process occurs via the magnetization rotation within the areas $2R_c$. In fields $H > H_R$, the magnetization dispersion in the samples obeys the Akulov's law

$$d_{\rm m} = \frac{1}{2} \left(\frac{D^{1/2} H_{\rm a}}{H} \right)^2,$$
 (3)

where H_a is the effective (observed in the magnetization curve) local-anisotropy field acting within the radius R_c (that is taken to be equal to the grain radius). In fields $H < H_R$, the magnetization curve is described by the function

$$d_{\rm m} = \frac{1}{2} \frac{\left(D^{1/2} H_{\rm a} \right)^2}{H_R^{3/2} H^{1/2}},\tag{4}$$

which indicates the existence of stochastic domains in samples. In fields $H < H_R$, the exchange interaction causes the formation of stochastic domains, the radius of which progressively increases from R_c to R_L as the field decreases from H_R to $H \rightarrow 0$. Thus, two parameters, such as $D^{1/2}H_a$ and H_R can be determined directly from the magnetization curve. The effective localanisotropy field H_a (Table 4) can be estimated assuming the uniaxial symmetry of randomly oriented easy axes of local anisotropy (D = 1/15).

In accordance with the theory of correlation magnetometry [6], the contribution of effective localanisotropy field to the root-mean-square fluctuation of anisotropy field of a stochastic domain $D^{1/2}\langle H_a \rangle$ can be estimated, using the experimentally determined $D^{1/2}H_a$ and H_R value, as

$$D^{1/2} \langle H_{\rm a} \rangle = \frac{\left(D^{1/2} H_{\rm a} \right)^4}{H_R^3}.$$
 (5)

The relative radius of a stochastic domain R_L/R_c can be estimated by the equation

$$\frac{R_L}{R_c} = \left(\frac{H_R}{D^{1/2}H_a}\right)^2.$$
 (6)

Table 4 shows the obtained characteristics.

It follows from the random magnetic anisotropy model [3, 6] that, in the case of random distribution of easy magnetization axes of stochastic domains, the coercive force of the material is $H_c \approx D^{1/2} \langle H_a \rangle$. When the easy magnetization axes of the stochastic domains are parallel, the hysteretic properties are determined by field $\langle H_a \rangle$. It can be seen from a comparison of $\langle H_a \rangle$, $D^{1/2} \langle H_a \rangle$, and H_c magnitudes (Table 4) that the $D^{1/2} \langle H_a \rangle$ field makes the main contribution to the coercive force of the Fe₉₅Zr₅ and Fe₈₅Zr₅N₁₀ films. For the Fe and Fe₉₀N₁₀ films, the $\langle H_a \rangle$ field makes the main contribution to the coercive force ($M_r/M_s > 0.5$ indicates the orientational coherence of magnetizations in the majority of stochastic domains in the absence of external magnetic field).

Saturation Magnetostriction

The magnetostriction curves $\lambda(H)$ of the studied films were measured in fields up to 80 Oe (the curves are given in [9]). To estimate the magnetoelastic anisotropy by equation $K_{\rm ME} = (3/2)\lambda_{\rm s}\sigma$, the saturation magnetostriction $\lambda_{\rm s}$ must be available.

The dependence $\lambda(H)$ is a function of the M(H)curve due to the similarity of the autocorrelation properties of the magnetostriction and magnetization. It is known for microcrystalline ferromagnets [11] that the saturation magnetostriction λ_s , like the saturation magnetization M_s , can be determined by the Akulov's saturation approach law, by the extrapolation of the $\lambda(H)$ dependence to field $H = \infty$ ($H^{-2} = 0$). We suggest describing magnetostriction in fields $H < H_R$ (for the studied films, H_R exceeds fields used in [9], which are less than 80 Oe) by the equation

$$\lambda = \lambda_{\rm s} \left[1 - \frac{1}{235} \frac{2}{H_R^{3/2} H^{1/2}} \right]. \tag{7}$$

Here, 2/35 is the dispersion of magnetostriction– anisotropy axes [11]. The experimental magnetostriction curves [9] were measured under reversible demagnetization conditions; therefore, the theoretical dependence (Eq. (7)) can be applied for them. Note that the suggested Eq. (7) was not derived theoretically strictly in terms of perturbation theory [6]. Because of this, coefficients in the dependence $\lambda \sim H^{-1/2}$ can be different. Figure 2 shows experimental data of Fe film and their fitting (solid line) by Eq. (7). Table 5 shows the saturation magnetostriction λ_s estimated by this procedure. The magnetostriction of the studied films is discussed in [9].

Components of Local Magnetic Anisotropy

The experimentally determined constants of the effective local magnetic anisotropy $K_{\rm eff} = H_{\rm a}M_{\rm s}/2$ (Table 6) substantially exceed the magnetocrystalline anisotropy constant (K_1) for Fe single crystal (~4.8 × 10⁵ erg/cm³ [10]), which comprises the natural magnetoelastic anisotropy ($\lambda_{\rm s}^2 E$, where *E* is the Young modulus). This difference between the $K_{\rm eff}$ and K_1 constants can be related to additional contributions of the magnetoelastic ($K_{\rm ME}$), surface ($K_{\rm a, s}$), and magnetostatic ($K_{\rm MS}$) anisotropies. To confirm the assumption, we quantitatively estimated the contributions from local magnetoelastic, surface, and magnetostatic energies.

The contribution of the magnetoelastic anisotropy to the local anisotropy was estimated by equation

$$K_{\rm ME} = \frac{3}{2} \varepsilon \lambda_{\rm s} \frac{E_{\rm f}}{1 + v_{\rm f}},\tag{8}$$

where ε is the microstrain of ferromagnetic phase (Table 2); $E_{\rm f}$ is the Young modulus of film (it is taken to be equal to 1.42×10^{12} erg/cm³ [12]); and v_f is the Poisson's ratio (is taken to be equal to 0.3). Table 6 shows the obtained $K_{\rm ME}$ magnitudes.

X-ray diffraction analysis showed the presence of the Fe_2N phase in the nitrogen-containing films. According to the Kersten's inclusion theory [5], magnetic poles must arise on the surface of Fe₂N grains located in the ferromagnetic matrix, which cause the local magnetostatic anisotropy $K_{\rm MS} = (3/2) M_{\rm s}^2 V_{\rm Fe_2N}^{2/3}$, where V_{Fe_2N} is the volume fraction of the Fe₂N phase (Table 2). Table 6 shows the obtained $K_{\rm MS}$ quantities. The assumption about the existence of the magnetostatic anisotropy is substantiated by the fact that the Fe₂N-phase grain size (that is 78 and 35 nm for the Fe₉₀N₁₀ and Fe₈₅Zr₅N₁₀ films, respectively) is higher than the critical size $D_{\rm cr} = 2(A/2\pi M_{\rm s}^2)^{1/2}$ [13] (where A ~ 10^{-6} erg/cm is the exchange interaction stiffness), which is ~6 nm for the studied films. When the grain size is less than the critical size, the exchange interaction almost completely suppresses magnetization inhomogeneities, in particular caused by demagnetizing fields.



Fig. 2. Magnetostriction of Fe film as a function of $H^{-1/2}$.

Grain boundaries of the Fe₂N and α -Fe phases are areas characterized by a broken symmetry of atomic arrangement on the surface of ferromagnetic α -Fe grains. The surface magnetic anisotropy [4] of the films is estimated by the equation

$$K_{\rm s} = \frac{3}{9\sqrt{2}}a\lambda_{\rm s}\frac{E_{\rm f}}{1+\nu_{\rm f}},\tag{9}$$

where *a* is the lattice parameter of the α -Fe phase (Table 2). The contribution of the surface anisotropy to the local anisotropy field is

$$K_{\rm a,s} = \frac{6K_{\rm s}V_{\rm Fe_2N}}{2R_{\rm Fe_2N}},$$
 (10)

where $2R_{Fe_2N}$ is the Fe₂N-phase grain size (Table 2). Table 6 shows the obtained K_s and $K_{a,s}$ quantities. Note that Eq. (10) was derived based on the assumption that surfaces characterized by the surface anisotropy K_s are spherical; for this surface, the $K_{a,s}$ anisotropy is zero due to the equivalence of all axes of the sphere. The consideration of spherical grains with Eq. (10) is possible since the model, which is used to process the X-ray diffraction pattern, determines the grain size assuming their spherical shape.

After subtracting the $K_{\rm ME}$, $K_{\rm MS}$, and $K_{\rm a, s}$ contributions from the $K_{\rm eff}$ quantity (Table 6), it becomes close to the magnetocrystalline anisotropy costant for bcc iron, which is ~ 4.8×10^5 erg/cm³. Thus, it can be seen that the high constants of the effective local magnetic anisotropy for the studied Fe-based films most likely comprise the $K_{\rm ME}$, $K_{\rm MS}$, and $K_{\rm a, s}$ contributions.

Table 5. Saturation magnetostriction (λ_s) of the studied films

Film	Fe	Fe90N10	Fe ₉₅ Zr ₅	$\mathrm{Fe}_{85}\mathrm{Zr}_5\mathrm{N}_{10}$
λ_s , 10 ⁻⁶	17 ± 3	-8 ± 2	7.5 ± 0.5	21 ± 2

Sample	Fe	Fe ₉₀ N ₁₀	Fe ₉₅ Zr ₅	$\mathrm{Fe}_{85}\mathrm{Zr}_5\mathrm{N}_{10}$	
$K_{\rm eff}$, 10 ⁵ erg/cm ³	10.1 ± 3.4	12 ± 2.9	6.4 ± 0.8	10.5 ± 0.7	
K_{ME} , 10 ⁵ erg/cm ³	1.6 ± 0.3	0.7 ± 0.2	0.28 ± 0.03	0.83 ± 0.32	
K_{MS} , 10 ⁵ erg/cm ³		3.5 ± 0.5		4.2 ± 0.4	
K_S , erg/cm ²	0.139 ± 0.025	0.067 ± 0.017	0.062 ± 0.004	0.18 ± 0.02	
$K_{a, S}, 10^5 \mathrm{erg/cm^3}$		0.13 ± 0.04		0.67 ± 0.08	
$K_1, 10^5 {\rm erg/cm^3}$	4.8 (magnetocrystalline anisotropy constant of bcc Fe [10])				

Table 6. Components of effective local magnetic anisotropy $K_{\rm eff}$

Table 7. Parameters of macroscopic magnetic anisotropy

Film	$H_{\rm c}$ Oe	$_{\sigma}H_{\rm c}$, Oe	$V_1, \%$	<i>V</i> ₂ , %
Fe	13 ± 2	220	87	13
$Fe_{90}N_{10}$	90 ± 5	245	92	8
Fe ₉₅ Zr ₅	45 ± 5	146	66	34
$Fe_{85}Zr_5N_{10}$	40 ± 5	127	65	35

Components of Macroscopic Magnetic Anisotropy

We assume that, in the case of partial suppression of effective local anisotropy by exchange interaction, it is the magnetoelastic anisotropy that makes the main contribution to the coercive force. In this case, we can estimate the coercive force of films by equation ${}_{\sigma}H_{c} = (3/2)\lambda_{s}\sigma/M_{s}$ using measured values of macrostresses (σ from Table 3) and saturation magnetostrictions (λ_{s} from Table 5). The obtained ${}_{\sigma}H_{c}$ vquantities (Table 7) substantially exceed the coercive force H_{c} determined from the hysteresis loops (Tables 4 and 7). This fact confirms indirectly the above statement that the H_{c} magnitudes for the studied films are determined by the properties of stochastic domains.

It is known [14] that, in the case of two main sources of the macroscopic magnetic-anisotropy energy with comparable volume fractions, the dM/dH

10

stim let H_c 0.110 H_c 0.110 H_c 0.1100 H_c 0.1100 H_c 0.1

Fig. 3. Field dependence of differential susceptibility dM/dH of the Fe₈₅Zr₅N₁₀ film.

differential susceptibility curve exhibits two peaks. The dM/dH curve for the studied films is described adequately by the sum of two Lorentz functions with peaks corresponding to the magnetic fields H_c and $_{\sigma}H_c$ as follows (Fig. 3):

$$\frac{dM}{dH} = \frac{2V_1}{\pi} \frac{\omega_1^2}{4(H - H_c)^2 + \omega_1^2} + \frac{2V_2}{\pi} \frac{\omega_2^2}{4(H - \sigma H_c)^2 + \omega_2^2},$$
(11)

where ω_1 and ω_2 are the integral widths of the peaks and V_1 and V_2 are the peak areas. The V_1 and V_2 fractions in the film volume (Table 7) have substantially different magnetic anisotropy energies (associated with H_c and ${}_{\sigma}H_c$, respectively). The fairly adequate coincidence of the estimated ${}_{\sigma}H_c$ quantities (Table 7) with the position of second peak in the dM/dH curve (Fig. 3) indirectly confirms the magnetoelastic nature of magnetic anisotropy of the V_2 volume fraction. The V_2 volume fraction with the magnetic anisotropy that determines the ${}_{\sigma}H_c$ quantity has the smaller autocorrelation radius in the film plane compared to that of the V_1 volume fraction with the magnetic anisotropy determining the H_c magnitude. This follows from equation $R_L = R_c (H_R/D^{1/2} \langle H_a \rangle)^{1/2}$, which, for the V_1 and V_2 volumes, differs only in the $D^{1/2} \langle H_a \rangle$ quantity that is proportional to H_c or ${}_{\sigma}H_c$ for the corresponding volumes V_1 or V_2 , respectively.

Based on the aforesaid, we can conclude that the magnetic anisotropy in volume V_1 , which determines the coercive force, is caused by the suppression of local anisotropy by exchange interaction (that couples grains in stochastic domains), whereas the magnetic anisotropy for volume V_2 , which affects the hysteresis loop shape, is determined by magnetoelastic energy from macrostresses.

The magnitudes of relative remanence M_r/M_s (Table 4, Fig. 4) can be explained as follows. The magnetostriction (λ_s) and macrostresses (σ) in the Fe and Fe₉₀N₁₀ films have the same sign. This increases the squareness of hysteresis loops (easy magnetization axes of magnetoelastic anisotropy are in the film plane). The magnetostriction (λ_s) and macrostresses (σ) in the Fe₉₅Zr₅ and Fe₈₅Zr₅N₁₀ films are opposite in



Fig. 4. Hysteresis loops for the $Fe_{90}N_{10}$ and $Fe_{85}Zr_5N_{10}$ films.

sign. This fact results in the substantial volume fraction of the perpendicular anisotropy (magnetic anisotropy of V_2) and $M_r/M_s < 0.5$. It is important to note that, in contrast to the saturation magnetization and coercive force, the relative remanence M_r/M_s is affected by the demagnetizing factor of a sample. The demagnetizing factor in the film plane is $N \le 7h_f/l$, where h_f is the film thickness and l is the film length along the magnetic measurement direction [15]. The maximum demagnetizing field for the studied films is $H_N = -NM_s$ and does not exceed -0.6 Oe, which is several times lower than the error of measurements for points in the hysteresis loop.

CONCLUSIONS

The nanocrystalline Fe, $Fe_{95}Zr_5$, $Fe_{90}N_{10}$, and $Fe_{85}Zr_5N_{10}$ films prepared by magnetron sputtering have been studied. The experimentally determined effective local magnetic anisotropy of the films is considered to be the sum of contributions from the magnetocrystalline, magnetoelastic, magnetostatic, and surface anisotropies. It is shown that the shape of hysteresis loops of the studied films is determined by the existence of two main components of macroscopic magnetic anisotropy; one of them is caused by stochastic domains determining the coercive force, while the other is related to the magnetoelastic anisotropy due to internal residual macro stresses and magneto-striction, which affect the shape of the hysteresis loop.

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