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Microwave methods as a means to estimate the uniformity of the magnetic parameters of multilayer structured elements

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The possibility of microwave methods (ferromagnetic and spin-wave resonance) for characterize of multilayer planar elements having modulation of both structural and magnetic parameters of the systems is demonstrated. The measurements carried out in out-of-plane orientation allowed us to detect the range of orientation angle of the applied magnetic field, within which the system can be considered as an effective medium with a small dispersion of average dimensions. Also the analysis of the angle dependences allowed us to estimate a number of the fundamental magnetic parameters: effective magnetization, exchange interaction constant, surface anisotropy constant, the field of perpendicular magnetic anisotropy.

Keywords: ferromagnetic and spin-wave resonance, multilayer two-dimesional elements, magnetic anisotropy, weak magnetic field sensors.

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Introduction

Metallic multilayer materials with nanoscale layers demonstrate a variety of effects that are in demand in various high-tech industries: giant magnetoresistance [1,2], interlayer exchange interaction [3–5], perpendicular magnetic anisotropy [5–8], anomalous Hall effect [19], strengthening of Dzyaloshinskii-Moriya interphase interactions [10] and so on. A wide range of properties is achieved by varying the elemental composition of the individual layers, their thickness and quality of the interface. The development of multilayer systems that use magnetic materials, the properties of which can be modified when exposed to external magnetic field (its direction, magnitude, frequency) is still a relevant task today.

The main technological advantage of multilayer films is the availability of a stable technology to fabricate multilayer structures and film elements based on them [11,12]. But there is a certain ambivalence in relation to the properties they exhibit. One of the main effects observed in such structures is the exchange bond between ferromagnetic layers, which, on the one hand, largely determines the observed unique phenomena of the multilayer structures. On the other hand, its presence may lead to a compromised unambiguity of the system's response as a sensor to external impact. Thus, the procedure for the synthesis of multilayer systems requires a tool (method) for assessing the degree of the system homogeneity in terms of its magnetic parameters.

Dynamic (MW) methods — ferro-magnetic resonance (FMR) and spin wave resonance (SWR) — are required when estimating a wide range of magnetic parameters. They are used in determining the fundamental parame-

ters of magnetic materials: effective magnetization $M_{\rm eff}$, exchange interaction constant A, spin-wave stiffness η , surface anisotropy constant K_S . The angular dependences of the parameters of FMR and SWR spectra (positions of resonant fields, intensity) at different orientations of the external permanent magnetic field, as well as the analysis of the spectrum structure, make it possible to determine the presence and contribution of anisotropy (magnetically crystallographic, magnetically elastic, surface) [13-16], interlayer exchange interaction [17], type of surface anisotropy [18,19]. The success of dynamic methods in characterizing multilayer structures is demonstrated by the results of both theoretical [20-24] and experimental [25-31] studies. The main findings obtained for the multilayer structures are the dependence of the spectrum shape on both the structural (thickness of the layers and their number) and magnetic parameters of the individual layer (magnitude of magnetization, field of anisotropy) [31].

The purpose of this work is to study multilayer planar elements [Cu/FeNi]₅/Cu/FeNi/Cu/FeNi/Cu/[Cu/FeNi]₅ having modulation of both structural and magnetic parameters of the system, using dynamic methods to evaluate the limits within which the system can be considered as an efficient environment with a small variance of the average parameters.

1. Samples and experimental methods

 $\begin{array}{lll} Film & structures & [Cu(3~nm)/Fe_{20}Ni_{80}(100~nm)]_5/\\ Cu(150~nm)/Fe_{20}Ni_{80}(10~nm)/Cu(150~nm)/Fe_{20}Ni_{80}(10nm)/\\ Cu(150~nm)/[Cu(3~nm)/Fe_{20}Ni_{80}(100~nm)]_5 & were & obtained \end{array}$

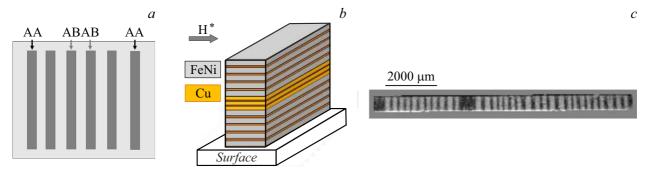


Figure 1. Scheme illustrating the geometry of a multi-layered element: H^* — process magnetic field applied during sputtering (a and b). Features of magnetic domain structure of an element, visualized using MOKE microscopy (c).

in argon medium using the system of radiation ion-plasma sputtering (Orion 8 (AJA InternationalInc., USA) at pre-vacuum of 1.3 · 10⁻⁶ mbar and operating pressure of Argon of $3.9 \cdot 10^{-3}$ mbar. Cover glasses with a thickness of $0.2 \, \text{mm}$ (Corning, $22 \times 22 \, \text{mm}$) were used as substrates. The film structures were obtained using an external process field H^* with a strength of 250 Oe applied in the sample plane to form an induced uniaxial magnetic anisotropy. Structuring with copper layers was used to obtain magnetic media of high thickness while maintaining high magnetic permeability and low coercivity [32]. For the same purpose, additional structuring of the central conductive copper layer was carried out, since it was previously found that in the absence of this layer the multilayer structures with similar magnetic properties are practically unfeasible when they are located before and after the conductive copper layer [26].

6 rectangular film elements parallel to each other $(10 \times 0.5\,\mathrm{mm}$ each) were obtained from a continuous multilayer film structure deposited on a single substrate by method of ion plasma sputtering in an additional single process cycle using optical lithography. The elements were oriented parallel to each other and in such a way that the direction of application of the external technological field coincided with the short side of the element (Fig. 1,a). Figure 1 shows a general structural view of a multilayer element and an image of the element magnetic domain structure obtained using magneto-optical Kerr effect (Evico, Dresden, Germany), confirming that a planar uniaxial magnetic anisotropy is formed along the short side of the element.

Two multilayer elements — AA and AB were selected for magnetodynamic studies, having the same process parameters (layer thickness, number of layers, alloys of individual layers and their sequence), but differing in position relative to the active sputtering zone of the target. Although the process parameters for fabricating the multilayer films (the ratio of the size of the target and the substrate, the distance to the target) ensured uniformity of physical and chemical characteristics of the film structure, the processes used during lithography could result in some variability in the properties of various elements. These differences could especially relate to the elements located at the edge

(elements of type AA) and in the center (elements of type AB) of the formed series.

Microwave spectra of films were made using equipment of the Krasnoyarsk Regional Center of Research Equipment of Federal Research Center "Krasnoyarsk Science Center of the Siberian Branch of the Russian Academy of Sciences" (spectrometer ELEXSYS E580, Bruker, Germany). Microwave spectra were measured at a room temperature in Xrange (resonator pumping frequency of $f=9.48\,\mathrm{GHz}$), the sample was placed into antinode of variable magnetic field h_{\sim} of the cavity resonator. The microwave absorption curves were decomposed into components using the differentiated Lorentz function. Measurements were carried out when permanent magnetic field changed its direction in angle θ_H (out-of-plane geometry of experiment as illustrated in Fig. 2).

The resonance frequency ω_0 of FMR [15,16,33] may be expressed via the total energy of magnetic system based on Landau-Lifshitz equation for the motion of magnetization M:

$$\omega_0 = \frac{\gamma}{M \sin \theta} \left[\frac{\partial^2 E}{\partial \theta^2} \frac{\partial^2 E}{\partial \varphi^2} - \left(\frac{\partial^2 E}{\partial \theta \partial \varphi} \right)^2 \right]^{1/2}, \tag{1}$$

where $\gamma = 1.758 \cdot 10^7 \, \text{Hz/Oe}$ — hyromagnetic ratio, θ and φ — polar and azimuth angles of magnetization in the spherical coordinate system, respectively.

The equilibrium position of magnetization vector is determined by the following relation:

$$\frac{\partial E}{\partial \varphi} = \frac{\partial E}{\partial \theta} = 0, \tag{2}$$

while the density of free energy [15]:

$$E = -MH \left[\sin(\theta) \sin(\theta_H) \cos(\varphi - \varphi_H) + \cos(\theta) \cos(\theta_H) \right]$$

+
$$[2\pi M^2 + K_n]\cos^2(\theta) + K_u\sin^2(\theta)\sin^2(\varphi - \varphi_0),$$
 (3)

where K_n — constant of perpendicular uniaxial anisotropy; K_u — constant of uniaxial anisotropy in the plane, φ_0 — angle characterizing the direction of uniaxial anisotropy field in the plane. Since the film is nanocrystalline, contributions of cubic anisotropy are excluded.

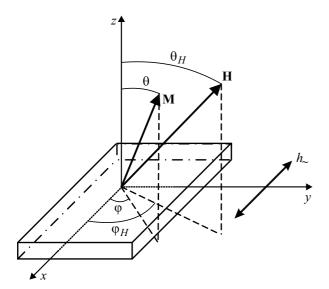


Figure 2. Scheme illustrating the geometry of the experiment.

2. Experimental findings and discussion

AA film has been certified by both static and dynamic magnetic methods [34]. The magnetic characteristics measured by different methods had a fairly good qualitative and quantitative consistency. In this paper, we used microwave methods to compare two planar elements in terms of the uniformity of their magnetic parameters. AA and AB films MW absorption spectra, measured at outof-plane orientation had a composite structure. Examples of experimental curves for $\theta_H = 90^{\circ}$ AA and AB films are shown in Fig. 3, a, c, respectively. The experimental curves of AA film in the angle range $90 > \theta_H > 40^\circ$ can be described by 4 Lorentzians (Fig. 3, a), AB films— 3 Lorentzians in the angle range $90 > \theta_H > 7^{\circ}$ (Fig. 3, c). The angular dependences of the resonance fields of the individual modes of AA and AB films are shown in Fig. 3, b, d, respectively. The several peaks in FMR spectrum of a multilayer planar structure, as we believe, is observed due to the implementation of effective layers that do not have an exchange coupling with each other [35]. The difference in the number of layers of multilayer films (10, ferromagnetic alloy layers) from the number of peaks detected in FMR spectrum (4 or 3, identifiable modes) may be due to the fact that a separate effective layer is formed by several ferromagnetic layers. Further analysis of the experimental spectra was performed under the condition of uniformity of the magnetic parameters within the effective layer.

The angular dependences of the selected modes were calculated from the system of equations (1)-(3), provided that the anisotropy field in the plane $(2K_u/M_S)$ 37 and 4 Oe was the same for the elements AA and AB, respectively, the effective magnetization $M_{\rm eff}$ was assumed to be 880 G for the individual effective layers of each element. The values of anisotropy field in the plane were obtained from the

Table 1. Fitting values of anisotropy field

$M_{\mathrm{eff}} = 880\mathrm{G}$	Perpendicular anisotropy field of individual magnetic phase H_{an} , Oe			
	1	2	3	4
AA AB	414 500	1000 100	1200 100	5000

angular dependence of the resonant field when the constant magnetic field changes in angle φ_H (Fig. 2 and [34]). The difference M_{eff} of a single-layer permalloy film from the magnetization value of a multilayer film with a Cu-interlayer having a negligibly small thickness (3 nm) relative to the thickness of the permalloy layer (100 nm), according to our estimate, is no more than 5%. This assumption is confirmed by the difference between the magnitude of the resonant field of a single-layer film (960 Oe) and the values of the resonant fields of individual modes of FMR spectra of AA and AB elements (for example, the modes of the microwave curve of element AB have resonant fields 946, 970 and 1000 Oe). Therefore, we consider it possible to use $M_{\rm eff}$ to describe the angular dependences of resonant fields, the value of which was estimated from FMR curve at $\theta_H = 0^\circ$ for a single-layer permalloy film with a thickness of 100 nm [30]. The variable parameter for obtaining a satisfactory match between the calculated curve and the experimental values of the resonant field was the magnitude of the perpendicular anisotropy field $(H_{an} = 2K_n/M_S)$. The data of theoretical matching were considered satisfactory with the divergence between the experimental and calculated values of no more than 5 and 1% for AA and AB respectively. The values found by this fitting H_{an} , are given in Table 1. It should be stressed that accuracy in determination of H_{an} in similar way were proved by other methods [30,36].

A distinctive feature of the angular dependences of the resonance fields of homogeneous modes (FMR) of multilayer AA and AB films is the range of angles within which individual homogeneous modes are observed. For AA film it was $90 > \theta_H > 40^\circ$, while for AB the range was $90 > \theta_H > 7^\circ$, which is significantly higher and similar to ranges observed for the one-layer films [37]. We assume that the larger size of the angle range within which homogeneous modes are realized for AB film stands for smaller fluctuations in magnetic parameters across the thickness of the effective layer within which the homogeneous mode is realized. Smaller values of the perpendicular anisotropy field (Table. 1) may also be due to the fact that each effective layer of the multilayer film AB had a more homogeneous magnetic structure.

The experimental spectrum in perpendicular geometry $(\theta_H = 0^\circ)$ for each film has a structure that can be divided into separate regions — A, B, C and D (Fig. 4, a, e). Each region is described by its own set of standing exchange modes (Fig. 4, b, f), the resonant fields H_n of which are described

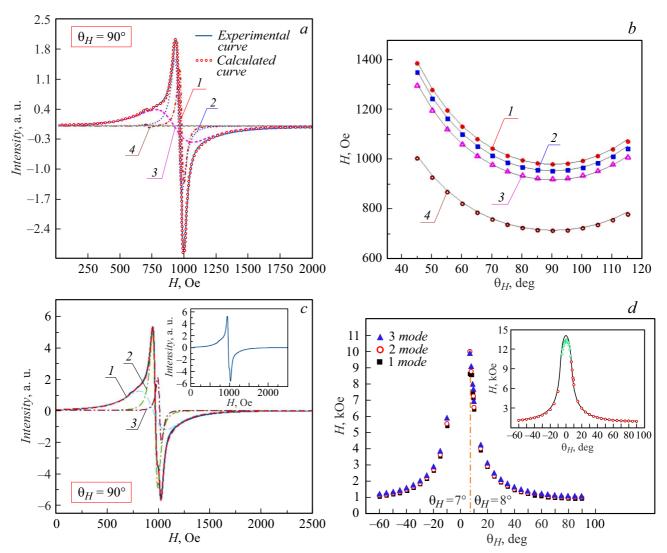


Figure 3. An example of experimental microwave absorption spectra for AA (a) and AB (c) films, as well as the angular dependences of the resonant fields of individual homogeneous modes (FMR) for AA (b) and AB (d). The solid lines on the fragment (b) represent the fitting curves. The fragment (c) shows experimental curve without resolution into separate modes. The insert of the fragment (d) shows the combination of experimental values (markers in the form of a circle and a triangle) with the calculated curve (solid line), the green triangles represent the resonant fields of the first standing exchange mode. Digits l-4 in the figure denote individual homogeneous modes.

with a high degree of accuracy by linear dependence on the square of the mode number (Fig. 4, with, d) and can be analyzed using the expression [37,38]:

$$H_n = \frac{\omega_0}{v} + 4\pi M_{\text{eff}} - \eta_{\text{eff}} k^2, \tag{4}$$

where $\eta_{\rm eff}=2A/M_S$ — spin-wave stiffness related to the exchange interaction constant A, M_S — saturation magnetization, $k=\pi n/d$ wave vector depending on the mode number n and the film thickness d.

The SWR spectra of AB film are significantly better resolved, which is explained by the smaller line width ΔH of the individual standing modes. Minimal values ΔH for AA film makes 40 Oe, while for AB film — 25 Oe. The

smaller dimensions ΔH of the AB film also indicate a more uniform structure of each individual effective layer.

Individual regions in SWR spectrum of AB film, which are recorded at $-6 < \theta_H < 6^{\circ}$, can be correlated with individual FMR peaks which are realized in the range $90 > \theta_H > 7^{\circ}$ (Fig. 3, d). The SWR spectrum regions of AA film cannot be compared with the detected FMR modes, since at values θ_H from 40 to 0° , the spectra are represented by both homogeneous magnetization fluctuations in one region and standing exchange spin modes excited in another region. The latter assumption reflects a significant difference in the formed boundary conditions of each effective layer.

Thus, measurements in *out-of-plane* geometry allow us to conclude that there are several effective layers in a planar multilayer composite, each of which is characterized

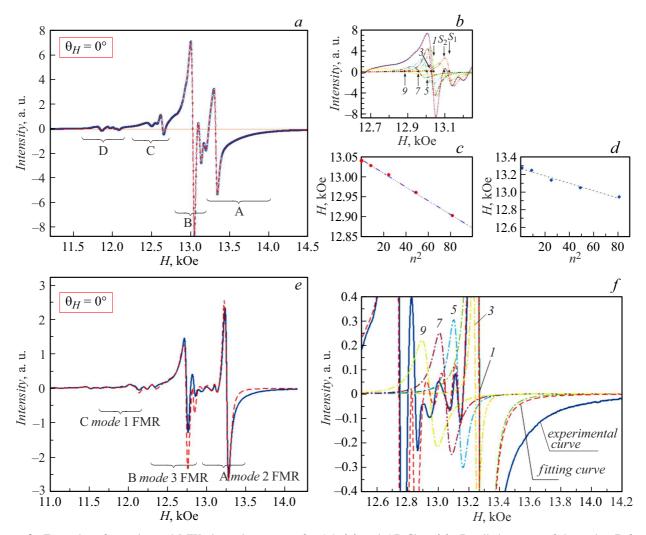


Figure 4. Examples of experimental MW absorption spectra for AA (a) and AB films (e). Detailed spectra of the region B for film AA (b) and region A for film AB (f) are presented with separate standing exchange modes indicated by Arabic numerals and dotted lines. (c) and (d) show the dependences of the resonant field on the square of the mode number for AA and AB films, respectively. The observed surface modes in region B for AA film(b) are denoted by S_1 and S_2 .

by its own set of magnetic parameters (internal field, perpendicular anisotropy field) [39]. The detection of the spin-wave resonance spectrum in the perpendicular geometry in experiment and the linear dependence of the resonant fields positions on the square of the mode number (Fig. 4, c) indicates that there's a sufficient homogeneity of magnetic parameters distribution across the thickness of each effective layer [29]. The last conclusion was made on the basis of studies [24–32], where it was shown that inhomogeneities in magnetic parameters usually lead to various deviations from the quadratic law of dispersion.

Using the expression (4), the exchange stiffness in field coordinates $\tilde{\eta}_{\text{eff}}$ and

$$\tilde{\eta}_{\text{eff}} = (H_1 - H_n)/(n^2 - 1)$$
 (5)

surface anisotropy constant were estimated.

$$|K_S| = \left[\frac{M_{\text{eff}}A}{2}\left[(H_S - H_1) - \frac{2A}{M_{\text{eff}}}\left(\frac{\pi}{d}\right)^2\right]\right]^{1/2},$$
 (6)

where H_S and H_1 — resonance fields of the surface and first standing mode, respectively (Fig. 4). The numerical values of $\tilde{\eta}_{\text{eff}}$ and presented in Table 2 (where K_{S1} and K_{S2} are the constants of surface anisotropy on individual surfaces of the effective layer, which are detected using the corresponding surface modes S_1 and S_2 of the spectrum at $\theta_H = 0^\circ$, an example of identification of S_1 and S_2 is shown in Fig. 4, b).

The difference of $\tilde{\eta}_{\rm eff}$ of the considered laminated films from the similar value of the single-layer permalloy film (50 Oe with a film thickness of 100 nm), as well as high values of H_{an} (table. 1) indirectly confirm the proposed model of the effective layer, which is formed by alternating ferromagnetic (FeNi) and nonmagnetic (Cu) layers. We believe that lower values H_{an} for AB element demonstrate a more uniform distribution of magnetic parameters within the effective layer. The effective exchange with such a planar composite structure is explained by two contributions — partial exchange of the ferromagnetic layer and partial

Table 2. The exchange stiffness and the constant of the surface anisotropy of individual effective layers within which a standing exchange spin wave is formed

Element	Magnetic phase	$ ilde{\eta}_{ ext{eff}},$ Oe	$ K_{S1} $, erg/cm ²	$ K_{S2} $, erg/cm ²
AA	A	5	0.017	0.014
	В	1.5	0.035	0.042
	С	9	_	_
AB	A	6	_	_
	В	11	0.025	_
	С	4	_	_

exchange between the ferromagnetic layers through a non-magnetic interlayer [25,40].

Conclusion

Multilayer planar elements [Cu/FeNi]₅/Cu/FeNi/Cu/ FeNi/Cu/[Cu/FeNi]₅ having non-periodic modulation of both structural and magnetic parameters in thickness were investigated by ferromagnetic and spin-wave methods resonances. The microwave spectra of both samples have a composite structure. The recorded FMR curves are represented as separate peaks, and the SWR curves are given as three separate regions, in each of which a set of standing exchange spin waves is detected. We consider individual peaks in FMR spectra and exchange-correlated regions in SWR as excitation of magnetization from several effective layers, each of which is characterized by its own set of magnetic parameters (internal field, perpendicular anisotropy field). Individual regions in SWR spectrum of AB sample are better resolved, which indicates more uniformly distributed magnetic parameters across the effective layer thickness and may serve as the basis for selecting proper element for a technological application.

The values of such fundamental magnetic parameters as effective magnetization, exchange interaction constant, and the surface anisotropy constant were estimated from the characteristics of microwave spectra at $\theta_H=90$ and 0° . The analyzed angular dependences of resonance characteristics in the *out-of-plane* geometry enabled us to determine the ranges of external magnetic field orientation angles, within which the system may be considered as an effective medium with a small variance of average parameters, as well as a field of perpendicular magnetic anisotropy.

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Conflict of interest

The authors declare that they have no conflict of interest.

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