DOI 10.1007/s11182-019-01673-4

Russian Physics Journal, Vol. 61, No. 12, April, 2019 (Russian Original No. 12, December, 2018)

PHYSICS OF MAGNETIC PHENOMENA

TWO-MAGNON RELAXATION PROCESSES IN NANOCRYSTALLINE THIN MAGNETIC FILMS

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UDC 537.621; 004.942

Numerical analysis of the micromagnetic model was used to reveal the 'resonance' feature of relaxation processes in nanocrystalline thin magnetic films. This feature manifests itself in the form of sharp broadening of the ferromagnetic resonance (FMR) line at a certain frequency f_1 depending on magnetic characteristics of the film, and is observed only in films the thickness of which exceeds some threshold value d_{min}. Sharp broadening of the FMR line is accompanied by significant shift of the resonance field, whereas the shift value changes the sign at frequency $\sim f_1$. It was shown analytically that the nature of observed effects is associated with the two-magnon process of spin waves scattering on quasi-periodic magnetic microstructure – magnetization 'ripple'. Obtained expressions for the threshold value of film thickness d_{min} and frequency of maximum broadening of FMR line f_1 agree well with the results of numerical computation of micromagnetic model.

Keywords: micromagnetic simulation, nanocrystallites, random magnetic anisotropy, ferromagnetic resonance, microwave frequencies, two-magnon relaxation process.

INTRODUCTION

It is known that thin magnetic films (TMFs) are widely used as a medium for recording of digital information on hard drives, however there are current studies aiming to create magnetic random-access memory on films [1]. In high-frequency magnetometers of weak magnetic fields, TMFs are used as sensing elements [2, 3], they are also the main elements in spintronics devices [4]. Maximum achievable parameters of such devices on TMFs are determined by dynamic magnetization characteristics that directly depend on relaxation processes. That is why a lot of attention is presently paid to studying the relaxation mechanisms and possibilities of controlling the relaxation processes in thin magnetic films [5].

It is obvious that the width of ferromagnetic resonance (FMR) line of any magnetic material is primarily determined by its intrinsic damping coefficient. However, there are also other extrinsic mechanisms of magnetization relaxation, among which the dominant contribution into high-frequency power absorption and broadening of FMR line in thin magnetic films is made by the processes of two-magnon scattering [6]. These processes are accompanied by damping of spin waves (magnons) during interaction with non-uniform internal magnetic fields that can emerge in magnetic medium for different physical reasons. For instance, the authors of [7, 8] study the effect of random local anisotropy on relaxation in polycrystalline thin films, and the authors of [9, 10] study the effect of random distribution

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of inhomogeneities and roughnesses on the surface of films. There are also studies focusing on the possibilities of controlling magnetic relaxation in TMFs by creating artificial magnetic inhomogeneities in them [11–14].

In thin nanocrystalline films, due to exchange interaction and magneto-dipole interaction of crystallites, magnetic moments form a wave-like quasi-periodic structure with a period depending both on magnetic film parameters and on the value of the applied external field [15, 16]. This non-uniform magnetic microstructure called the 'ripple' of magnetization increases damping of spin waves and can lead to a significant broadening of FMR line, as well as a resonance field shift, which was first shown by Ignatchenko and Degtyarev [17]. The goal of the present paper is to study the effect of non-uniform magnetic microstructure on relaxation in nanocrystalline thin films, using a micromagnetic model for numerical computation of the high-frequency susceptibility [18, 19].

1. NUMERICAL MODELING

For the sake of simplicity, research was done on films that are monolayers of densely packed nanoparticles of D_0 size with random orientation of anisotropy axes. The focus was on the micromagnetic model of film with thickness of $d = D_0$ and area of 1024×1024 discrete cells, the size of which corresponded to the size of nanoparticles and varied within the range of 12–100 nm. Analytical expression from [20] was used to calculate the tensor component of demagnetization coefficients conditioned by magneto-dipole interaction between nanoparticles. Two-dimensional periodic boundary conditions were applied to exclude edge effects associated with inhomogeneity of internal magnetic field near the boundaries of samples, when calculating the energies of exchange and magneto-dipole interactions [21].

For the sake of certainty, magnetic parameters of studied samples were selected in accordance with the well-known nanocrystalline alloy $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ [22], the saturation magnetization of which is $M_s = 955$ G ($\mu_0 M_s$ = 1.2 T), the exchange stiffness constant is $A = 1\cdot10^{-6}$ erg/cm ($1\cdot10^{-11}$ J/m), the damping parameter of spin waves is $\alpha = 0.005$, and the field of local uniaxial anisotropy is $H_k = 2K/M_s = 171.7$ Oe ($K = 8200$ J/m^3). However, it was assumed that the magnetic anisotropy of the entire film is absent. External static and alternating magnetic fields were applied in the film plane and were directed orthogonally to each other. Numerical implementation of the method of undetermined coefficients for solving the linearized system of Landau-Lifshitz equations was used to calculate high-frequency magnetic susceptibility of films [18].

As was already noted, magnetic inhomogeneities associated with the stochastic magnetic microstructure have a strong effect on the spectrum of high-frequency absorption of nanocrystalline thin films. Their effect leads not only to the shift of FMR field and broadening of the resonance line, but also to emergence of shape asymmetry of the resonance curve [18]. It is obvious that resonance field H_R and line width ΔH of FMR obtained by numerical computation of micromagnetic model can be written in the form of sums

$$H_R = H_0 + H_{2\mathrm{m}},$$
$$\Delta H = \Delta H_0 + \Delta H_{2\mathrm{m}}, \tag{1}$$

where the first terms of the right part of each of the two expressions characterize the resonance line of uniform FMR of the film in the absence of non-uniform magnetic microstructure, while the second terms show the shift of the resonance field $H_{2\mathrm{m}}$ and broadening of FMR line $\Delta H_{2\mathrm{m}}$ as a result of two-magnon scattering of spin waves on inhomogeneities. The resonance field H_0 satisfies the equation $\omega_0 = 2\pi f_0 = \gamma\sqrt{H_0(H_0 + 4\pi M_s)}$, where $\gamma = 1.76\cdot10^7$ rad/(s·Oe) is gyromagnetic ratio. The line width of uniform FMR is determined by the well-known expression $\Delta H_0 = 4\pi\alpha f_0/\gamma$.

According to the theory of two-magnon relaxation processes developed by Arias and Mills for ultra-thin films [9], the frequency dependence of the broadening of FMR line $\Delta H_{2\mathrm{m}}$ is described by the expression

$$\Delta H_{2\mathrm{m}} = \Gamma \arcsin\frac{H_0}{H_0 + 4\pi M_s} = \Gamma \arcsin\sqrt{\frac{\sqrt{f_0^2 + (f_M/2)^2} - f_M/2}{\sqrt{f_0^2 + (f_M/2)^2} + f_M/2}}, \tag{2}$$

TABLE 1. Dependencies on Film Thickness of the Frequency of Maximum FMR Line Broadening f_1 due to Two-Magnon Relaxation Mechanism on Magnetization 'Ripple', the Value of Broadening ΔH_{2m}, as well as the Resonance Field H_0 and the Line Width ΔH_0 for Films in the Absence of 'Ripple'

d, nm	f_1, GHz	ΔH_{2m}, Oe	H_0, Oe	ΔH_0, Oe
24	10.75	16.86	1121	38.4
32	6.65	25.74	452	23.7
42	4.66	35.01	225	16.6
56	3.46	47.6	129	12.5
75	2.89	82.2	89	10.4
100	3.26	123.7	111	11.6

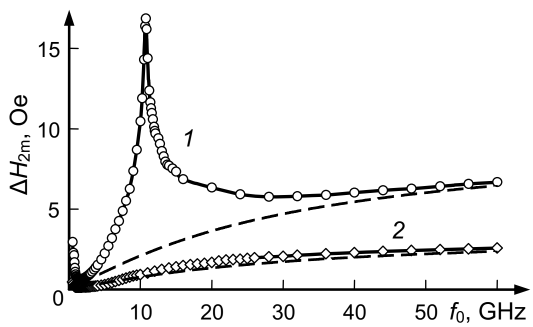

Fig. 1. Frequency dependencies of FMR line broadening ΔH_{2m} obtained by numerical micromagnetic simulation of the high-frequency susceptibility of nanocrystalline thin films with thickness of d =12 nm (*1*) and 24 nm (*2*). Dashed lines are dependencies according to the formula (2).

widely used in practice to interpret experimental dependencies $\Delta H(f_0)$. Here $f_M = 2\gamma M_s$, and Γ is coefficient characterizing 'intensity' of magnetic inhomogeneities. As one can see from (2), the dependence $\Delta H_{2m}(f_0)$ is a function monotonously increasing until some saturation without any features.

However, as numerical computation of the micromagnetic model of TFM showed, in the case of nanocrystalline films with a thickness over some threshold value at a certain frequency f_1 depending on film parameters, one observes a sharp increase in high-frequency power absorption and as a consequence – a sharp broadening of FMR line. As an example on Fig. 1 for two values of the film thickness $d = 12$ and 24 nm solid lines indicate the dependencies $\Delta H_{2m}(f_0)$ obtained by numerical computation of the high-frequency susceptibility of nanocrystalline thin films. Dashed lines indicate the dependencies built according to the formula (2). One can see that for the film with thickness of $d = 12$ nm the theory put forward by Arias and Mills describes rather well the obtained dependence $\Delta H_{2m}(f_0)$. However, for the film with thickness of 24 nm the broadening of FMR line $\Delta H_{2m}(f_0)$ has a sharp peak at frequency $f_1 \approx 10.75$ GHz, whereas at high frequencies one observes a rather good agreement of micromagnetic computation with the theory by Arias and Mills.

With an increase in film thickness, the revealed 'resonance' feature of two-magnon relaxation is maintained, whereas the relaxation contribution ΔH_{2m} to FMR line width quickly increases and the 'resonance' frequency f_1 monotonously decreases (Table 1). From the table one can see that for the 24 nm thick film, due to two-magnon relaxation processes, the width of FMR line increases approximately 1.5 times, while for the 100 nm thick film it increases by more than an order of magnitude. One should note that with the increase in film thickness the asymmetry of FMR line shape increases in a monotonous manner. As one expected, the broadening of FMR line by means of the two-magnon mechanism of magnetization relaxation by value ΔH_{2m} is simultaneously accompanied by a significant change in resonance field by value H_{2m} in relation to field H_0 corresponding to the field of film FMR without

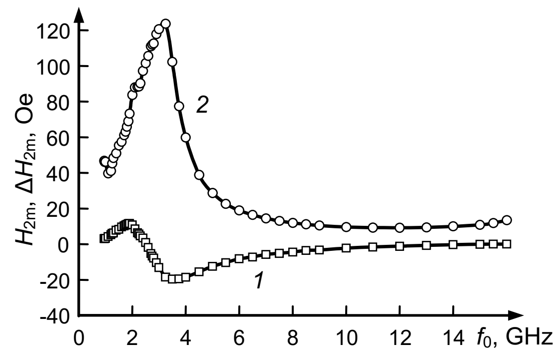

Fig. 2. Frequency dependencies of the resonance field shift H_{2m} (curve *1*) and the broadening of FMR line ΔH_{2m} (curve *2*) obtained by numerical micromagnetic simulation of high-frequency susceptibility of the 100 nm thick nanocrystalline film.

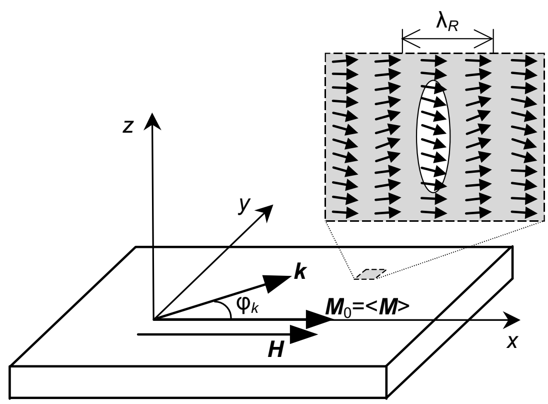

Fig. 3. Model of thin magnetic film and schematic image of longitudinal magnetization 'ripple'. Magnetic correlated region is marked with an ellipse.

magnetization 'ripple'. This fact is confirmed by the dependence $H_{2m}(f_0)$ presented in Fig. 2. One can see that with the increase in frequency f_0 addition to the resonance field has a positive sign, but somewhere around frequency f_1 = 3.26 GHz it changes the sign, while the dependence $H_{2m}(f_0)$ has two extreme points.

2. THEORETICAL MODEL

To explain the nature of revealed effects associated with behavior of magnetic relaxation in nanocrystalline thin films, the authors considered the two-magnon model of spin waves scattering on magnetic inhomogeneities emerging due to non-uniform magnetic microstructure in TMF – magnetization 'ripple'. It is important to note that the main distinctive feature of nanocrystalline thin magnetic films is small size of crystallites compared to the effective radius of exchange and magneto-dipole interaction [23]. That is why the presence of the magnetic coupling between crystallites leads to averaging and partial suppression of the random magnetic anisotropy of individual crystallites. However, such suppression of the local anisotropy usually is not complete, which leads to deviations of the magnetization vector $\boldsymbol{M} = M_s\boldsymbol{m}$ in relation to some average direction $\boldsymbol{M}_0 = \langle \boldsymbol{M} \rangle$. This is why a peculiar magnetic structure emerges with spatial deviations of magnetization near some average direction (Fig. 3), called magnetization 'ripple' in research literature [15, 16].

The most rigorous and consistent static theory of such fine magnetic microstructure was developed by Hoffmann [15]. Hoffmann, based on electronic microscopy results, developed a model of non-interacting between each other magnetic correlated regions formed in the film. The size and shape of such regions depend on the radius of the exchange and magneto-dipole interaction, on the crystallite size and the value of applied magnetic field. In the general case such region coupled by magnetic interaction (magnetic correlated region) is an ellipsoid strongly stretched in the direction perpendicular to direction of the average magnetization $\boldsymbol{M}_0$ (Fig. 3). In the linear approximation [15], the length of ellipsoid semi-axis ($R_{||}$) along the average direction of magnetization is $R_{||} = \sqrt{D/H}$, where $D = 2A/M_s$.

Using averaging of the magnetic anisotropy of individual crystallites, with a random orientation of anisotropy axes, within the magnetic correlated region, Hoffmann derived an expression for dispersion of the transverse component of magnetization, as well as the most probable main period length of longitudinal magnetization 'ripple':

$$\lambda_R = 2\pi R_{||} = 2\pi\sqrt{D/H}\ . \tag{3}$$

Quasi-periodic magnetization structure with a period λ_R forms quasi-periodic magnetic fields of demagnetization with the same period in the thin nanocrystalline film. This is equivalent to formation in the film of magnetic inhomogeneities with a characteristic 'size' $R_{||}$ and wave number:

$$k_R = 2\pi/\lambda_R = 1/R_{||} = \sqrt{H/D}\ . \tag{4}$$

A distinctive feature of these inhomogeneities is that their size depends not only on magnetic parameters of the film, but also on the value of applied external field.

Within the theory of two-magnon relaxation processes [6, 7, 9, 24, 25], magnetic inhomogeneities, including those that are also associated with non-uniform stochastic magnetic structure, are considered as perturbation of intrinsic magnetic oscillations (spin waves) of the uniform sample. Inhomogeneities violate orthogonality of intrinsic magnetization oscillations and lead to coupling between them. This causes energy transfer from the considered type of oscillations (for instance, uniform FMR) into non-uniform types of oscillations, i.e. to emergence of additional dissipation, as well as shift of resonance frequencies [6, 7, 9].

Let us write dispersion equation of spin waves for the model of uniform thin magnetic film (Fig. 3) [9]:

$$\omega_k = \gamma\sqrt{[H + Dk^2 + 4\pi M_s N_k][H + Dk^2 + 4\pi M_s \sin\varphi_k (1 - N_k)]}\ . \tag{5}$$

Here Dk^2 is the field of exchange interaction for a spin wave with the wave vector $\boldsymbol{k}$ ($k = |\boldsymbol{k}|$), H is the value of planar external magnetic field coinciding with the direction of equilibrium magnetization $\boldsymbol{M}_0$, φ_k is the angle between the direction of spin wave propagation and equilibrium magnetization $\boldsymbol{M}_0$, N_k is the demagnetizing factor that depend on the wave number k. In approximation of thin film, magnetization of which insignificantly changes throughout its thickness, this factor is of the form [16]

$$N_k = \frac{1 - e^{-kd}}{kd}\ . \tag{6}$$

Dispersion dependence (5) is graphically represented on Fig. 4. Here ω_0 shows the frequency of uniform FMR with $k = 0$, and curves with $\varphi_k = 0$ and 90° respectively represent the lower and upper boundaries of the spin wave spectrum. One can see that dispersion curves at $\varphi_k < \varphi_k^{\text{crit}}$ cross the line ω_0. This means that the frequency of uniform FMR coincides with frequencies of the group of spin waves, for which $0 < k_i < k_{\max}$, while the presence of a non-uniform internal magnetic field (magnetic inhomogeneities) with the wave number coinciding with k_i ensures the transfer of energy of uniform thin film excitation into spin wave energy with the wave number k_i. The so-called two-magnon scattering of spin waves on inhomogeneities occurs.

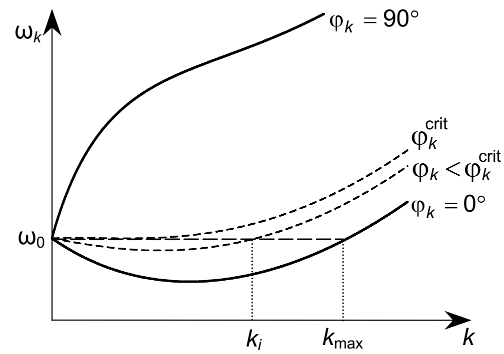

Fig. 4. Dispersion dependencies of spin waves for different directions of their propagation φ_k.

The maximum wave number of degenerate states k_{max} is determined from the condition

$$\omega_k(H, k_{max}, \varphi_k = 0) = \omega_0 . \tag{7}$$

It is obvious that the largest scattering of spin waves on magnetic inhomogeneities of magnetization 'ripple' with the wave number k_R (see formula (4)) will be observed upon the condition of equality $k_{max} = k_R$. That is why the field value H_1, at which maximum of FMR line broadening will be observed, is determined from the condition

$$\omega_k(H_1, k_R, \varphi_k = 0) = \omega_0 . \tag{8}$$

To find the analytical solution to this equation, it is necessary to simplify the expression for demagnetization factor N_k determined by formula (6). The authors of classical publications [7, 9] on the theory of two-magnon processes in thin films use an approximation for ultra-thin films $kd \ll 1$, according to which $N_k \approx 1 - kd/2$. However, as will be shown below, the scattering of spin waves on inhomogeneities of stochastic magnetic structure is possible only for films having thickness over a certain threshold value. Whereas the value $k_R d$ is of order 1, and N_k is better approximated with the expression $N_k \approx 1 - kd/e$. Then equation (8) turns to quadratic equation in relation to variable $x = \sqrt{H_1}$:

$$\frac{3}{4\pi M_s} x^2 - \frac{2d}{e\sqrt{D}} x + 1 = 0 . \tag{9}$$

From the condition of the existence of solution to equation (9) minimum film thickness

$$d_{min} \approx \frac{e}{2}\sqrt{\frac{3D}{\pi M_s}} \approx 1.33\sqrt{\frac{D}{M_s}} , \tag{10}$$

at which the effect of magnetization 'ripple' on two-magnon relaxation processes becomes a determining factor. Minimum thickness d_{min} for the used magnetic parameters of numerical model, according to (10), is $d_{min} \approx 19.7$ nm. The resulting value conforms well to the numerical simulation results presented in Fig. 1.

One should note that equation (9) has an accurate solution, but its approximate formula has the simplest analytical form:

$$x = \sqrt{H_1} \approx \frac{e}{2}\frac{\sqrt{D}}{d} . \tag{11}$$

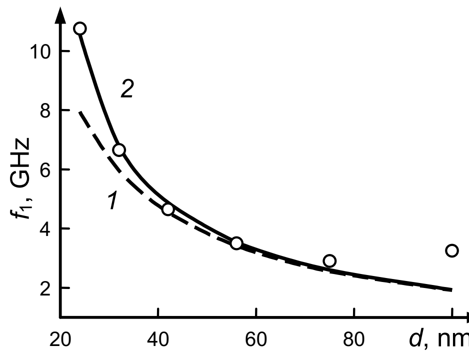

Fig. 5. Dependence of frequency $f_1 = \omega_1/2\pi$, at which one observes the maximum broadening of FMR line, on the thickness of nanocrystalline thin magnetic film. Circle markers are the result of numerical computation of micromagnetic model, curve *1* is calculation according to approximate formula (12), curve *2* is accurate solution to equation (9).

Directly from (11) and taking (4) into account we get the value $k_R d \approx 1.36$, as well as expression for resonance frequency ω_1, at which the maximum broadening of FMR line will be observed:

$$\omega_1 = \gamma\sqrt{H_1(H_1 + 4\pi M_s)} \approx \gamma\frac{e}{d}\sqrt{\pi M_s D} = \gamma\frac{e}{d}\sqrt{2\pi A}\,. \tag{12}$$

On Fig. 5 the theoretical dependence $f_1(d) = \omega_1(d)/2\pi$ obtained with the help of approximate formula (12) is indicated by dashed curve *1*, while solid curve *2* reflects the results of accurate solution of equation (9). One can see that the biggest difference between accurate and approximate solutions is observed near the threshold film thickness $d_{\min}$. On the same figure circle markers indicate dependence $f_1(d)$ obtained on the basis of numerical computation of micromagnetic model that shows good agreement with an accurate solution to equation (9).

However, one should note the emergence of an increasing difference of computational experiment results (computation of micromagnetic model) and solution to equation (9) for 'thick' films with $d > 80$ nm. For such films the maximum broadening of FMR line is observed at relatively low excitation frequencies, that is in the region of small magnetic fields H (see table). In this case the linear approximation used by Hoffmann when deriving the expression for λ_R (3) for the film located in small magnetic fields becomes too rough. Meanwhile, it is obvious that in the theoretical model it is necessary to take into account the impact of non-linear terms, when calculating internal non-uniform magnetic fields considerably increasing in weak external fields due to increase in magnetization dispersion in the film [15, 16].

CONCLUSIONS

In this paper, the effect of magnetic microstructure of magnetization 'ripple' on two-magnon relaxation processes in nanocrystalline thin magnetic films was studied. Numerical computation of the high-frequency magnetic susceptibility using a micromagnetic model of TMF showed that magnetization 'ripple' exerts considerable impact on relaxation in nanocrystalline films, but only in the case of thicknesses exceeding some threshold value. In particular, it was established that the broadening of FMR line has a sharp peak at a certain frequency f_1 associated with the thickness and magnetic parameters of the film. Meanwhile, the considerable shift of resonance field of FMR is observed, and the shift value changes the sign at frequency $\sim f_1$. It is important to note that for films having thickness below the threshold value, the dependence of FMR line broadening on frequency shows monotonous increase to some saturation, and this dependence is well described by classical theory by Arias and Mills [9].

To explain the nature of revealed distinctive features of magnetic relaxation in nanocrystalline thin films, the authors performed an analytical calculation of the film model, where one takes into account two-magnon scattering of spin waves on magnetic inhomogeneities emerging due to non-uniform magnetic microstructure of magnetization 'ripple'. As a result, an expression was obtained for calculation of the threshold value of film thickness, above which the sharp broadening of FMR line is observed at a certain frequency f_1. The formula for calculation of the frequency f_1 was also obtained. The largest scattering of spin waves on magnetic inhomogeneities of magnetization 'ripple' is observed in the case of equality of wave numbers of the 'ripple' and spin waves, that is why the maximum broadening of FMR line occurs only at a certain frequency. Note that the main results of analytical calculation of the considered TMF model conform well to the results of numerical analysis of the micromagnetic model.

This research was done with support of the Russian Ministry of Education and Science, task No. 3.1031.2017/PCh.

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