SOLID MATTER

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RELAXATION AT NONLINEAR FERROMAGNETIC RESONANCE

Nonlinear ferromagnetic resonance in yttrium iron garnet films have been studied both experimentally and theoretically. It is shown that the application of Landau–Lifshitz equation with a relaxation term in the Gilbert form brings about a qualitatively incorrect result in the determination of the Q-factor for a magnetostatic microwave resonator, because the theory predicts a growth of the Q-factor, when the pump signal power increases to a critical value, above which the foldover phenomenon occurs. When using a modified relaxation term in the form of a power series in the time derivative of the magnetization, the results of calculations coincide with experimental ones to a sufficient accuracy. The nonlinearity term (coefficient) in the equation for the uniform precession is shown to depend on the fields of uniaxial anisotropy of the first and second orders. This fact can be used to affect the characteristics of nonlinear processes in ferromagnets.

K e y w o r d s: nonlinear ferromagnetic resonance, relaxation term, magnetic crystallographic anisotropy.

1. Introduction

To describe the dynamics of magnetization, the Landau–Lifshitz equation [1] with a relaxation term is applied. As a rule, the latter is used in the Gilbert form [2]. However, modern experiments [3–6] testify that the application of this term at large precession angles gives rise to a discrepancy with experimental data even at the qualitative level. There are at least a few ways to modify the relaxation term in the Landau–Lifshitz equation. In particular, in work [7], the method aimed at constructing the dissipative function and finding the relaxation term with regard for the type of crystal symmetry was developed. Another approach consists in representing the relaxation Gilbert term as a series expansion in the time derivative of the magnetization [6].

The relaxation substantially affects the precession with large deviation angles, so that a possibility for such nonlinear phenomenon as the bistability associated with the microwave power absorption and called the foldover [8–10] has to be taken into account. A necessary condition for this phenomenon to be observed is that the amplitude of a pumping microwave field should achieve a threshold value. The magnitude of threshold field was obtained theoretically in work [11] for an ellipsoid of revolution taking the shape anisotropy into account. However, the foldover threshold can depend not only on this parameter but also on the cubic crystallographic anisotropy [12].

This work aimed at the experimental verification of the nonlinear relaxation theory, which includes the modified relaxation term [6]. As an object of investigations, we used film specimens of yttrium-iron garnet (YIG) grown up on a substrate of gadolinium gallium garnet with the orientation (111). The researches concerned, in particular, a comparison of the dependences of the resonator Q-factor and the shift of the resonator resonance frequency on the pump power. In addition, the influence of the magnetic crystallographic anisotropy on nonlinear processes is

studied in the framework of three-parameter model with rgard for the cubic anisotropy fields and the uniaxial anisotropy of the first and second orders. The amplitudes of pumping fields in our researches were comparable with the threshold amplitudes needed for the foldover to start.

2. Landau–Lifshitz Equation with a Modified Relaxation Term in the Gilbert Form

The general form of the Landau–Lifshitz equation looks like

$$\frac{\partial \mathbf{m}}{\partial t} = -\gamma \left[\mathbf{m} \times \mathbf{H}^{\mathrm{eff}} \right] + \mathbf{R}, \tag{1}$$

where $\mathbf{m} = \mathbf{M}/M_0$ is the normalized magnetization, $\mathbf{H}^{\mathrm{eff}}$ an effective field defined as the variational derivative of the functional of the total ferromagnet energy with respect to the magnetic moment, γ the gyromagnetic ratio, and $\mathbf{R}$ the relaxation term [13]. The latter is taken in the form [6]

$$\mathbf{R} = \alpha \left(\eta \right) \left[\mathbf{m} \times \frac{\partial \mathbf{m}}{\partial t} \right], \tag{2}$$

where the function $\alpha \left(\eta \right)$ is given by the series

$$\alpha \left(\eta \right) = \alpha_{\mathrm{G}} \left(1 + q_1 \eta + q_2 \eta^2 + ... \right), \quad \eta = \frac{1}{\omega_M^2} \left(\frac{\partial \mathbf{m}}{\partial t} \right)^2,$$

α_{G} is the standard Gilbert relaxation constant, and q_i are empirical coefficients. Basing on the following estimation of η for YIG made by shifting the resonance frequency by the characteristic quantity $\Delta \omega = 10$ MHz:

$$\frac{(m^2)\omega^2}{\omega_M^2} = \frac{(1 - m_z^2)\omega^2}{\omega_M^2} = \frac{(2\Delta \omega)\omega^2}{\omega_M^3} = 4 \times 10^{-3},$$

the further consideration can be confined to the first two terms in the α-series.

Let us change in Eq. (1) to the circular variables $a_0 = m_x + i m_y$ and consider only the uniform precession of a magnetization. Let the uniform oscillations occur with the frequency ω, i.e. $a_0 \sim A e^{i\omega t}$. In the general case, this frequency can differ from the pump one, ω_p. The effective magnetic field is formed by the external magnetic bias field H_0, the circularly polarized transverse microwave pumping field h_0, and the demagnetizing field associated with the specimen

shape. Let the resonator have the disk shape. Then the diagonal tensor of demagnetizing coefficients $\hat{N}$ has the components N_T in the direction orthogonal to the rotation axis and the components N_Z in the parallel direction. For the specimen anisotropy to be made allowance for, the fields of the cubic anisotropy, H_c, and the uniaxial anisotropy of the first, H_{u1}, and second, H_{u2}, orders [14] were taken into consideration. Keeping the terms not higher than those of the third order in a_0, the nonlinear equation for the uniform precession written down in terms of circular variables looks like

$$\left[i + \alpha_{\mathrm{G}} \left(1 + \frac{q_1}{\omega_M^2} |\dot{a}_0|^2 \right) \right] \dot{a}_0 +$$
$$+ \left(\omega_0 + \sigma |a_0|^2 \right) a_0 = \gamma h_0 e^{i\omega_p t}, \tag{3}$$

where

$$\omega_0 = \gamma \left[H_0 + 4\pi M_0 (N_T - N_Z) - H_c + H_{u1} + H_{u2} \right],$$

$$\sigma = \gamma \left[\frac{4\pi M_0}{2} (N_Z - N_T) - \frac{1}{2} \left(H_{u1} + 3H_{u2} \right) \right].$$

Equation (3) is a partial case of the equation of motion for a magnetization in the S-theory [15], when the coupling between the uniform precession and spin waves, as well as between the spin waves themselves, is neglected. The account for spin waves in this problem should increase power losses for the uniform precession in comparison with the considered model and somewhat change the curves of resonance absorption.

In Eq. (3), the nonlinear coupling between the modes of the right and left circular polarizations is neglected. It can be done if

$$\left| \frac{\alpha_{\mathrm{G}} \omega_0}{4\pi M_0 \gamma (N_T - N_Z) - \gamma (H_{u1} + 3H_{u2})} \right| \ll 1.$$

As a rule, the anisotropy fields that enter the nonlinearity coefficient σ are lower than M_0 by an order of magnitude. If they are neglected, then, for a specimen in the form of a planar disk, the condition given above is reduced to $\frac{\alpha_{\mathrm{G}} \omega_0}{4\pi M_0 \gamma} \ll 1$. In our problem, this criterion is obeyed, so that the coupling between oscillators can be neglected.

First, let us obtain a solution for the homogeneous equation (3), i.e. let us determine the characteristic frequency of a nonlinear ferrimagnetic resonator. For this purpose, we use perturbation theory [16]. The

solution is sought as a series $a_0 = \sum_{j=1}^{3} \kappa^j a_0^{(j)}$ with the linear approximation $a_0^{(1)} = A e^{i\omega t}$ and the exact frequency value $\omega = \omega^{(0)} + \kappa\omega^{(1)} + \kappa^2\omega^{(2)}$, where κ is a small parameter, and $\omega^{(0)} = \omega_0 (1 + i\alpha_G)$ is the oscillation frequency in the linear approximation. In the higher-order approximations, we obtain the first and second frequency corrections, $\omega^{(1)} = 0$ and $\omega^{(2)} = [\sigma + i\alpha_G\omega_0 q_1 r]|A|^2$, respectively, where $r = \omega_0^2/\omega_M^2$.

In the linear approximation, the dependence of the amplitude of forced oscillations produced by a nonlinear oscillator on the amplitude and the frequency of an external force is given by the relation [16]

$$|A| = \frac{\gamma h_0}{\sqrt{\varepsilon^2 + \alpha_G^2\omega_0^2}}, \qquad (4)$$

where the detuning parameter $\varepsilon = \omega_p - \omega_0$ was introduced. If the correction $\omega^{(2)}$ to the resonance frequency ω_0 is made allowance for, Eq. (4) can be rewritten in the form

$$|A| = \frac{\gamma h_0}{\sqrt{(\varepsilon - \sigma|A|^2)^2 + \alpha_G^2\omega_0^2(1 + q_1 r|A|^2)^2}}.$$

By solving this quadratic equation for ε, we obtain the following two branches of the resonance curve:

$$\varepsilon_{1,2} = \sigma|A|^2 \pm \sqrt{\frac{\gamma^2 h_0^2}{|A|^2} - \alpha_G^2\omega_0^2(1 + q_1 r|A|^2)^2}. \qquad (5)$$

The threshold field amplitude, at which the foldover begins, can be determined from the condition that a vertical section appears in the dependence $\varepsilon(|A|)$,

$$h_{0\,th} = \frac{\Delta H B^2}{2\sqrt{2}} \sqrt{B^2\left[\rho^2 - 4(q_1 r)^2\right] - 4q_1 r}, \qquad (6)$$

where

$$B = \frac{8q_1 r + 2\sqrt{\rho^2 + 20(q_1 r)^2}}{\rho^2 + 4(q_1 r)^2},$$

$$\rho = \frac{2\sigma}{\alpha_G\omega_0},$$

and $\Delta H = \dfrac{2\alpha_G\omega_0}{\gamma}$ is the ferromagnetic resonance (FMR) line width.

The analysis of the threshold foldover fields for an ellipsoid of revolution was carried out in works [11, 13]. The threshold field values were obtained by
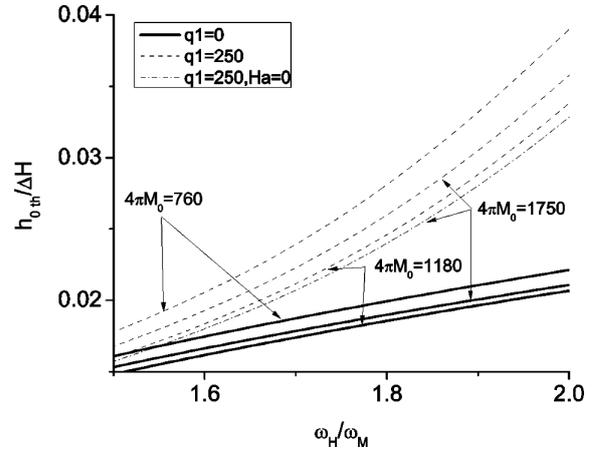
Fig. 1. Normalized dependences of the foldover threshold fields on the magnetic bias field for YIG and Ga-YIG specimens. The corresponding saturation magnetization values are indicated near the curves

exactly solving the Landau–Lifshitz equation with regard for the shape anisotropy

$$h_{0\,cr} = \frac{\Delta H}{\sqrt{2}\rho}\sqrt{\sqrt{\rho^2 + 1} - 1}.$$

At $q_1 = 0$, formula (6) corresponds to the result of works [11, 13], if $\rho \gg 1$; the latter condition being well satisfied for disks:

$$h_{0\,th} = \frac{\Delta H}{\sqrt{\rho}}.$$

Formula (6) makes it possible to plot the dependences of the threshold pumping field amplitude on the magnetic bias field for materials with various magnetic parameters. The range of bias fields was selected to satisfy the condition $q_1\eta \lesssim 1$. Pure and Ga-substituted yttrium iron garnets (Fig. 1) and barium hexaferrites (Fig. 2) fabricated in the disk form were studied. In the plots, the threshold field is normalized by the FMR line width.

The material parameters of Ga-substituted YIGs were taken from work [17], the parameters of hexaferrites from work [18], and the parameters of hexaferrite doped with scandium to $x = 0.3$ and 0.6 were obtained by interpolation. Figure 1 demonstrates the influence of a relaxation term in the Gilbert form and its modification, as well as the fields of magnetic crystallographic anisotropy, on the foldover threshold value. In the case of a modified relaxation term,
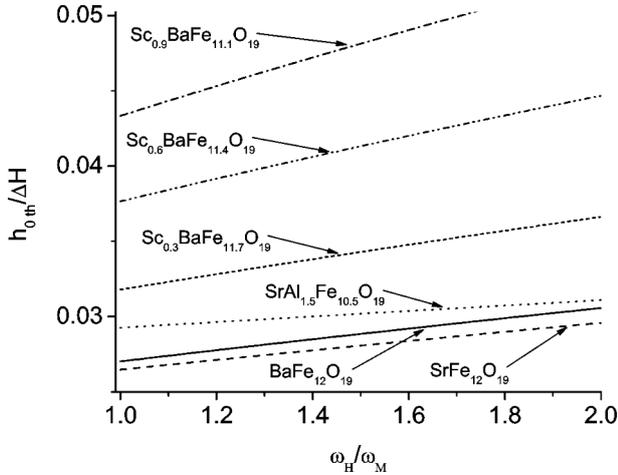
Fig. 2. Normalized dependences of the foldover threshold fields on the magnetic bias field for barium hexaferrites ($q_1 = 0$)

the critical field is higher, this result being in a better agreement with the experiment. The account for anisotropy fields also affects the foldover threshold value.

One can see from Fig. 2 that, in the case of hexaferrites doped with aluminum (the higher uniaxial anisotropy field), the normalized value of foldover threshold field changes insignificantly, whereas, at the doping with scandium (the anisotropy decreases), the threshold considerably grows.

From the Landau–Lifshitz equation, it follows that the nonlinearity coefficient σ in Eq. (3) depends on the fields of uniaxial anisotropy of the first and second orders. This fact is interesting for applications, because it assumes a capability of controlling the nonlinear properties of a ferromagnetic resonator. For instance, according to the estimations carried out in work [19], if an YIG film is used as a component of the planar two-layer structure ferrite/piezoelectric material, there exists a possibility to change the first-order uniaxial anisotropy field, H_{u1}, in the interval ± 20 Oe. This circumstance, in its turn, makes it possible to vary the value of nonlinearity coefficient within the limits of ± 28 MHz. The calculations show that the threshold amplitude for the foldover to start changes by 6% at that. For pump powers higher than 12 dBm, the theory predicts that the Q-factor should change by 4%, and the resonance frequency by no more than 5%.

3. Comparison between Theory and Experiment

In order to verify our theory, we experimentally determined the Q-factor for a nonlinear disk resonator 1.6 mm in diameter and 23 μm in thickness in the normal magnetic biasing mode with the bias field $H_0 = 3300$ Oe. The resonator was fabricated from a (111) YIG film with a saturation magnetization of 1750 Gs, and the anisotropy fields $H_c = -50$ Oe, $H_{u1} = -50$ Oe, and $H_{u2} = 45$ Oe. The measurement cell was a microstrip wave guide [14] that provided the linearly polarized microwave pumping to a magnetostatic resonator, the latter playing the role of an inhomogeneity in the transmission line. The relation between the amplitude of a microwave field and the transmitted power is

$$P = \frac{120 d^2}{\sqrt{\varepsilon_d}} \ln\left(\frac{r_B}{r_A}\right) h^2,$$

where ε_d is the dielectric permittivity of a transmission line material, and r_A and r_B are the roots of the transcendental equation

$$r - \ln r - 1 - \frac{\pi b}{2d} - \left(\frac{2\Delta}{d} + \sqrt{\frac{2\Delta}{d}}\right)(r - 1) = 0,$$

where the following notations are introduced: d is the thickness of the dielectric layer, and Δ and b are the thickness and the width, respectively, of the strip with the current. The magnetic field directed normally to the specimen surface was created by a dc NdFeB magnet; in such a way, field pulsations inherent to electromagnets were avoided. Under those conditions, oscillations of forward volume magnetostatic waves were excited in the ferrite resonator. In the course of experiment, the transmission coefficient of the microstrip line with the resonator was measured making use of a Rohde & Schwarz ZVA8 vector network analyzer.

It is known that, when studying nonlinear processes, the duration of a probing signal must be much longer than the characteristic time of establishment of a stationary mode for nonlinear oscillations, which is reciprocal to the relaxation frequency. We experimentally determined the required regime of measurements. For this purpose, we measured the dependence of the nonlinear frequency shift, $F_0^{(2)} - F_0^{(1)}$, of a magnetostatic resonator on the time of scanning

over the frequency (all other device tunings remained invariable). Here, $F_0^{(2)}$ and $F_0^{(1)}$ are the resonance frequencies measured at pump powers of $+15$ and -30 dBm, respectively. The corresponding results are shown in Fig. 3. One can see that the frequency shift saturates when the scanning duration exceeds 30 s. All experimental results reported in this work were obtained when the frequency scanning time was equal to 30 s, which corresponds to a scanning rate of 6.5 Hz/s and agrees with previous works [9, 10].

To compare the theoretical resonance curves with the experimental data, let us consider the nonlinear absorption at the FMR, which is associated with the microwave part of the Zeeman energy. In this case, the power losses $\mathrm{Im} \left(\frac{\omega_0}{2} M_0 \int h_0 a_0 dV \right)$ (see work [13]), provided that magnetization oscillations are uniform, can be written down in the form

$$P_a = V M_0 \frac{\alpha_\mathrm{G} \omega_0^2}{2\gamma} |A|^2 \left(1 + q_1 r |A|^2 \right).$$

Some examples of nonlinear absorption curves obtained experimentally are depicted in Fig. 4. The inclusion of the modified relaxation term (2) into the Landau–Lifshitz equation results in the frequency dependence of losses in the system, in contrast to the standard Gilbert model. The growth of power losses restricts the precession angle amplitude and reduces the Zeeman absorption of a microwave pump power at the resonance. The larger width of resonance curves obtained in the experiment can be explained by extra losses in the metallic surfaces of a wave guide, in the dielectric layer, and by reemission losses in a ferrite resonator. At the same time, the examined theoretical model makes allowance for only the intrinsic losses in ferrite, whereas the losses arising owing to parametrical processes and the interaction of spin waves with one another and also affecting the resonance width are omitted.

The nonlinear Q-factor of a resonator was determined experimentally following the technique described in work [20], namely, from the scalar amplitude-frequency characteristic of a resonator inserted into the transmission line as an inhomogeneity. The loaded Q-factor, Q_L, was determined from the formula $Q_L = F_0 / \Delta F$, where F_0 is the resonance frequency, and ΔF is the resonant linewidth corresponding to a half power absorption. The unloaded Q-factor, Q_0, is related to the loaded one by means of the coefficient K of resonator coupling with the wave
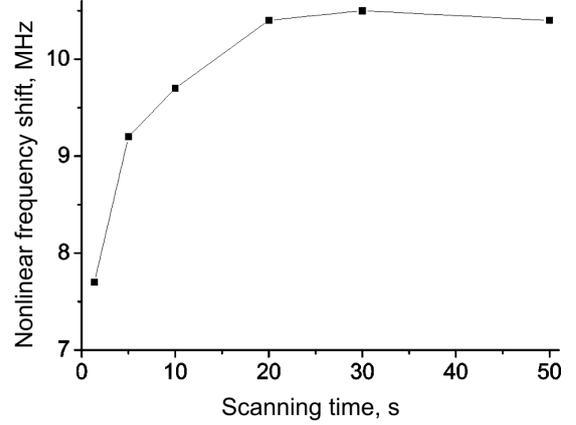
Fig. 3. Nonlinear shift of the magnetostatic resonator frequency as a function of the frequency scanning time
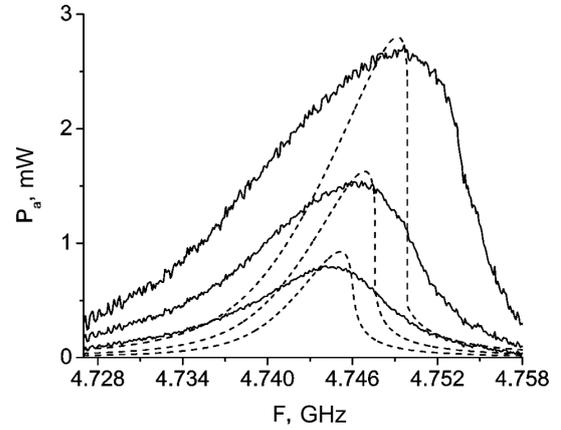
Fig. 4. Contours of energy absorption by the ferrimagnetic resonator at the pump power $P = 4$, 8, and 15.8 mW. Experimental (solid curves) and theoretical (dashed curves, $q_1 = 250$) results

guide, $Q_0 = (1 + K)Q_L$. At low levels of pump power, the unloaded Q-factor of a resonator was found to equal $Q_{0\,\mathrm{max}} = 1300$. This parameter was also calculated theoretically from the curves of resonance absorption (Fig. 4) using the formula $Q_0 = F_0 / \Delta F$. Formula (5) can be analyzed in two approximations: when only the first term proportional to α_G is taken into consideration in function (2) – this way corresponds to the application of a relaxation term in the Gilbert form; and when the modified relaxation term $\alpha(\eta) = \alpha_\mathrm{G} \left(1 + q_1 \dot{\mathbf{m}}^2 / \omega_M^2 \right)$ is used.

In the former case, the Q-factor of a nonlinear disk resonator grows with the pump power P_0 (Fig. 5, dashed curve). It should be noted that, although the
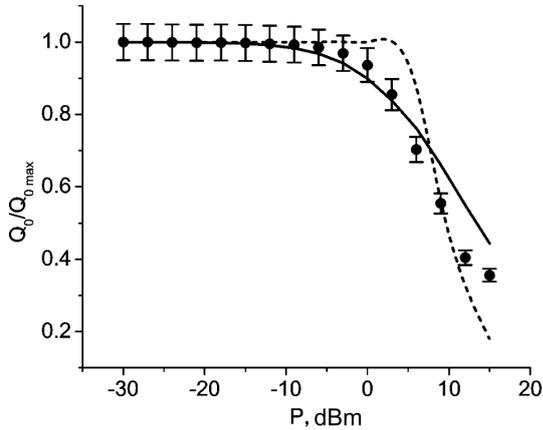
Fig. 5. Normalized unloaded Q-factor of a resonator ($Q_{0\,\max} = 1300$). Points correspond to experimental data; the dashed curve displays the result of calculations with a relaxation term in the Gilbert form ($q_1 = 0$), and the solid curve with the modified relaxation term ($q_1 = 250$)
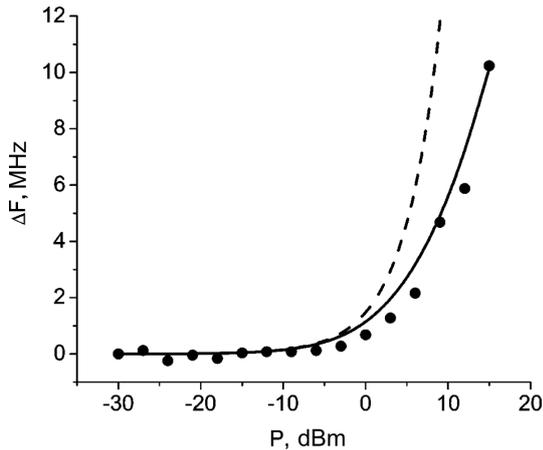
Fig. 6. Dependences of the resonance frequency shift on the pump power for a nonlinear magnetostatic resonator: (dashed curve) the results of theoretical calculations with a relaxation term in the Gilbert form, (solid curve) the results of theoretical calculations with the modified relaxation term ($q_1 = 250$), (points) experimental results

growth of Q-factor is insignificant, this result is physically incorrect even at the qualitative level. However, if the modified relaxation term is included into consideration and the first coefficient in the series expansion is put equal $q_1 = 250$, the behavior of the Q-factor corresponds to the experimental data (Fig. 5, solid curve).

The theory presented above also makes it possible to calculate the resonance frequency shift $\Delta F_0(P) =$

$= F_0(P) - F_0^{(1)}$ for various versions of a relaxation term and to compare the results obtained with experimental data (see Fig. 6). It is evident that the application of a modified relaxation term provides a good quantitative agreement between them.

At the qualitative level, the difference between two relaxation models can be explained as follows. The application of a relaxation term in the Gilbert form brings about lower power losses in comparison with the modified variant. As a result, the amplitude of magnetization oscillations and, accordingly, the resonance frequency shift are larger at identical values of pump power P. However, the experimental results testify in favor of the modified variant.

4. Conclusions

It is demonstrated that the application of a modified relaxation Gilbert term in the Landau–Lifshitz equation in the form of a series expansion and the proper choice of relevant parameters make it possible to explain, both qualitatively and quantitatively, the experimental data obtained for the unloaded Q-factor and the frequency shift of a nonlinear magnetostatic resonator. The value of empirical coefficient q_1 that provided the best agreement with the experiment is found. The equation for the nonlinear uniform precession is analyzed, and analytical expressions describing the resonance absorption curve for the nonlinear ferrimagnetic resonator are obtained. A possibility to control the nonlinearity coefficient by selecting the first- and second-order uniaxial anisotropy fields is pointed out.

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РЕЛАКСАЦІЯ ПРИ НЕЛІНІЙНОМУ
ФЕРОМАГНІТНОМУ РЕЗОНАНСІ

Р е з ю м е

Наведено результати експериментальних і теоретичних досліджень нелінійного феромагнітного резонансу в плівках залізо-ітрієвого гранату. Показано, що використання в рівнянні Ландау–Ліфшица релаксаційного члена в формі Гільберта дає якісно невірний результат при визначенні добротності магнітостатичного НВЧ-резонатора, адже в цьому випадку теорія передбачає зростання добротності при збільшенні потужності сигналу накачки до критичного значення, вище якого спостерігається явище фолдоверу. При застосуванні модифікованого релаксаційного члена, в формі ряду по ступенях похідної по часу від намагніченості, результати розрахунків з достатньою точністю збігаються з експериментом. Показано, що в рівнянні для однорідної прецесії коефіцієнти, що визначають нелінійність, залежать від полів одновісної анізотропії першого та другого порядків. Цей факт можна використати для впливу на характеристики нелінійних процесів в феромагнетиках.