MECHANISM OF ENERGY EQUILIBRIUM IN NONLINEAR FERROMAGNETIC RESONANCE

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A nonlinear detuning equilibrium mechanism due to variation of the crystal magnetization is taken into account in the microscopic theory of a strongly excited ferromagnetic spin system. It is shown that this mechanism predominates over the nonlinear dissipative mechanism considered in[1] or is commensurate with it and yields the Bloch model for magnetization relaxation. Experimental data confirming the theoretical conclusions are presented.

1. INTRODUCTION

THE state of a strongly excited spin system of a ferromagnet (beyond the threshold of parametric excitation) depends essentially on the magnetization relaxation mechanism. If the total magnetization is an integral of the motion in the relaxation process (the Landau-Lifshitz model), then a stationary state with constant amplitude and phase of the spin oscillations is established in the spin system. On the other hand, if the magnetization changes somewhat (the Bloch model), then slow undamped oscillations occur in the spin system, with a period on the order of the relaxation time[2]. Which of the aforementioned models can be applied to a strongly excited state is a question that can be answered by the microscopic theory of nonlinear ferromagnetic resonance (NFMR).

The quantum-mechanical theory of NFMR, describing the mechanism of dissipative limitation of the spin-wave growth, is developed in the paper of Suhl and Gottlieb[1] and is the development of the kinetic theory of many-magnon processes in ferromagnetic resonance (FMR)[3] as applied to the case of a strong radio-frequency field.

However, Suhl and Gottlieb did not consider the possibility of nondissipative limitation of the growth of the spin waves due to the automatic detuning of the natural frequency of the spin system, resulting from the change in the longitudinal component of the macroscopic magnetization M_z. Such a detuning lowers the efficiency of parametric excitation of the spin system, i.e., it decreases the influx of energy and by the same token leads to equilibrium.

We shall show below that a nonlinear detuning mechanism for the establishment of equilibrium between the energy supplied from the outside (from the pump) and the damping in a strongly excited spin system prevails almost everywhere over the mechanism of nonlinear dissipative limitation. The change of the demagnetization ΔM_z is described by the expression[4]

$$\Delta M_z = \frac{1}{V}\int M_z\,dV - M_0 = -\frac{\sum_k n_k}{N}\frac{\mu_B}{M_0 a^3}, \tag{1}$$

where n_k are the occupation numbers of the magnons, N the total number of atoms, V the volume of the crystal, a the lattice constant, and μ_B the Bohr magneton.

Owing to the dipole-dipole interaction, the natural frequency of the spin oscillations depends on the magnetization, and therefore the value of the latter causes the frequency to shift by an amount $\Delta\omega_k$ in proportion to the number of magnons:

$$\Delta\omega_k = 2\pi\gamma\frac{\omega_H'}{\omega_k}\Delta M_z \sin^2\theta_k, \tag{2}$$

where θ_k is the angle between the directions of the magnon quasimomentum (k) and the magnetization field (the latter is directed along the z axis),

$$\omega_H' = \omega_H + \omega_e(ak)^2, \quad \omega_H = \gamma H_0, \quad \omega_e = \gamma H_e, \quad \omega_M = 4\pi\gamma M_0,$$

H_0 is the magnetization field, H_e the exchange field, M_0 the magnetization per unit volume, γ the gyromagnetic ratio for the spin, and $\omega_k^2 = \omega_H'(\omega_H' + \omega_M \sin^2\theta_k)$ - natural frequency of the spin system.

2. NONLINEAR DISSIPATIVE AND DETUNING MECHANISMS

The number of magnons in a parametrically excited spin system of a ferromagnet is determined by the formula (see, for example,[1])

$$n_k = \frac{\lambda_k^2 + (\omega/2-\omega_k)^2 + p^2}{\{\lambda_k^2 + (\omega/2-\omega_k)^2 - p^2\}^2}|\alpha_k|^2, \tag{3}$$

where $1/\lambda_k$ is the transverse-relaxation time, ω the pump frequency, $|\alpha_k|^2$ the magnon-source intensity, $p = (\gamma h_0 \omega_M/2\omega_k)\sin^2\theta_k$, and h_0 is the amplitude of the pump field. For concreteness we are considering here the case of "longitudinal pumping," when the directions of the magnetization and pump fields coincide.

It is obvious that $n_k \to n_t$ as $p_k \to 0$, where n_t is the number of thermal magnons. Therefore formula (3) for beyond-threshold pumping can be rewritten in the form

$$n_k \cong 2n_t\left\{1 - \frac{p^2}{\lambda_k^2 + (\omega/2-\omega_k)^2}\right\}^{-2}. \tag{4}$$

The excitation threshold p_k is determined from the condition $n_k \to \infty$, i.e.,

$$p_k^2 = \lambda_k^2 + \left(\frac{\omega}{2} - \omega_k\right) \quad \text{or} \quad h_{0k} = \frac{2\omega_k}{\omega_M \sin^2\theta_k}\left\{\lambda_k^2 + \left(\frac{\omega}{2}-\omega_k\right)^2\right\}^{1/2} \tag{5}$$

and is minimal when the spin system is tuned to $\omega_k = \omega/2$.

It is known that mechanisms of nonlinear limitation of the growth of n_k are in operation in any real system beyond the excitation threshold. In particular, the nonlinear dissipative mechanism described by Suhl and Gottlieb[1] leads to a decrease in the total relaxation time λ_k', which can be written in the form

$$\lambda_k' = \lambda_k + \lambda_l(n_k), \tag{6}$$

where $1/\lambda_l$ is the time of "nonlinear relaxation," and l is an index corresponding to the number of magnons taking part in the interaction.

The nonlinear detuning mechanism leads to a shift of the natural frequency $\Delta\omega_k$ (see formula (2)). Substituting expressions (6) and (2) in (4) we obtain a nonlinear equation for the determination of the stationary value of n_k. Knowing the latter, we can calculate the characteristics of the NFMR, in particular the imaginary component of the magnetic susceptibility χ''[1], which can be readily monitored in experiment:

$$4\pi\chi'' = \frac{4\pi\Delta M_z''}{h_0} = \frac{1}{2(2\pi)^2}\frac{\gamma M_0}{p}\frac{\mu_B}{M_0 a^3}\frac{\omega_M V \sin^2\theta_k}{\omega_k N}\int n_k d^3k. \tag{7}$$

The indicated calculation program is based on the determination of the explicit functional dependence $\lambda_l(n_k)$. An exact solution of the latter problem is very cumbersome, but Suhl and Gottlieb developed a brilliant approximate method for calculating the functions $\lambda_l(n_k)$; we make use of their data[1)].

In NFMR, the region of the energy spectrum of the magnons with large n_k narrows down practically to a line. Thus, in the case of longitudinal pumping, magnons are excited along the spectral line $\theta_k = \pi/2$, and they make the main contribution to the energy dissipation. This makes it possible to simplify greatly the calculations, but at the same time lowers the probability of the collision processes, since it becomes more difficult to satisfy the energy and momentum conservation laws. The conservation laws are satisfied only in narrow regions of variation of the magnetizing field, for example for a three-magnon process ($l = 3$) in the region $0 < H_0 < H_{3m}$, for a four-magnon interaction ($l = 4$) in the region $H_{3m} < H_0 < H_{4m}$, etc., where

$$H_{3M} = \sqrt{(\omega/3\gamma)^2 + (2\pi M_0)^2} - 2\pi M_0,$$
$$H_{4M} = \sqrt{(8\omega/3\gamma)^2 + (2\pi M_0)^2} - 2\pi M_0.$$

Calculation shows[1] that the relaxation time of the foregoing processes increases like $\sim 10^l$. The more accurate values of the function λ_l are

1) $l = 3, \quad \lambda_3 = D_3 n_k$ for $0 < H_0 < H_{3M}$. (8)

($3\times10^{-2} < ka < 5\times10^{-2}$ in the case of yttrium garnet), where

$$D_3 \cong \frac{8}{\pi}\cos^2\theta_k \sin^2\theta_k \left(\frac{\omega_M}{\omega_e}\right)^2 \frac{p_k}{ka}\frac{\mu_B}{M_0 a^3}; \tag{8a}$$

2) $l = 4, \quad \lambda_4 = D_4 n_k^2$ for $H_{3M} < H_0 < H_{4M}$ (9)

($10^{-2} < ka < 3\times10^{-2}$ in the case of yttrium garnet), where

$$D_4 \cong \frac{2}{(2\pi)^4}\sin^2\theta_k \frac{\omega_M^2 p_k^2}{\omega_e^3}\left(\frac{\mu_B}{M_0 a^3}\right)^2; \tag{9a}$$

3) $l = 5$ (general case - in the entire region of existence of NFMR)

$$\lambda_5 = D_5 n_k^2, \tag{10}$$

where

$$D_5 \cong \frac{8}{(2\pi)^5}\cos^2\theta_k \sin^2\theta_k \frac{\omega_M^2\omega_k p_k^2}{\omega_e^4}(k'a)\left(\frac{\mu_B}{M_0 a^3}\right)^2, \tag{10a}$$

etc.

There is no need to write out the expressions for larger values of l, since we shall verify later that they are negligibly small compared with the detuning term. The latter can be obtained in a form convenient for comparison with expressions (8)–(10), by going over in (2) from the sum over k to an integral, in accordance with the substitution

$$\sum_k \to \frac{V}{(2\pi)^3}\int d^3k$$

and by using the aforementioned narrowing of the energy spectrum of the strongly excited spin system. The integration interval Δk is determined in this case by the expression

$$\Delta k \cong p_k / \omega_e a^2 k,$$

which can be readily obtained from the requirement that the width of the parametric excitation band be limited to the value $p - p_k$[1]. Taking these remarks into account, the detuning term can be written in the form

$$\omega/2 - \omega_k = R n_k, \tag{11}$$

where

$$R = \frac{1}{(2\pi)^2}\frac{\omega_H'}{\omega_k}\frac{\omega_M \sin^2\theta_k}{\omega_e}(ka)p_k\frac{\mu_B}{M_0 a^3}. \tag{11a}$$

It is convenient to characterize the comparative estimate of the contributions of the different equilibrium mechanisms by means of the parameter $\eta_l = f(D_l/R)$. It is easy to verify that its values are:

$$1)\ l = 3, \quad \eta_3 = \frac{D_3}{R} = 32\pi\cos^2\theta_k\frac{\omega_M}{\omega_e(ka)^2}, \tag{12}$$

$$2)\ l = 4, \quad \eta_4 = \frac{\sqrt{p_k D_4}}{R} = \sqrt{\frac{p_k}{\omega_e(ka)^2}} \ll 1, \tag{13}$$

$$3)\ l = 5, \quad \eta_5 = \frac{\sqrt{p_k D_5}}{R} \cong 20\,\mathrm{ctg}\,\theta_k\frac{\sqrt{\omega_k p_k}}{\omega_e\sqrt{ka}}\sqrt{\frac{\mu_B}{M_0 a^3}} \tag{14}$$

etc.

From an analysis of the threshold formula (5) it is known that the parameter ka changes with the magnetizing field like $\sqrt{(H_C - H_0)/H_e}$ when $H_0 < H_C$, where

$$H_C = \tfrac{1}{2}\{\sqrt{(\omega/\gamma)^2 + (4\pi M_0)^2} - 4\pi M_0\},$$

and when $H_0 > H_C$ the parameter ka remains, roughly estimated, of the order of 10^{-4}. Here $\theta_k \cong \pi/2$ when $H_0 \le H_C$ and decreases when $H > H_C$, causing an increase in the excitation threshold.

It is easy to see that $\eta_l < 1$ in almost the entire region of existence of the NFMR. Exceptions are two regions of the magnetization field: the first at H_0

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1) Suhl and Gottlieb use in their paper the explicit form of $\lambda_l(n_k)$ only for rough estimates (within 1–2 orders of magnitude), and frequently they replace the functional dependence of the parameters, for example θ_k, with numbers; we had to refine their data somewhat. This reduced to a more accurate substitution, into the kinetic equation, of coefficients that allow for terms of order 3, 4, and 5 with respect to the magnon creation and absorption operators in the Hamiltonian. These terms can be obtained, for example, in the paper of White and Sparks[5].

$\sim H_{3M}$, the second at $H > H_C$. In the first case we have ($ka = 3 \times 10^{-2}$ for yttrium garnet):

$$\eta_3|_{H_{3M}} \cong (15 \cos\theta_k)^2 \gtrsim 1 \quad \text{for } \cos\theta_k \sim 10^{-1}; \tag{15}$$

in the second case ($ka \sim 10^{-4}$):

$$\eta_3|_{H>H_C} \cong \operatorname{ctg}\theta_k. \tag{16}$$

Substituting expressions (8)–(11) in (4), we obtain nonlinear algebraic equations of third degree and higher for the determination of the stationary value of n_k. Their solution can be simplified by recalling that we are interested only in the largest real roots. Then, taking into account the estimate of the parameter η_l, we can easily find that[2)]

$$n_k = \frac{(2\pi)^2}{\sin^2\theta_k}\frac{p}{p_k} f\left(\frac{p}{p_k}\right)\frac{\omega_k}{\omega_H'}\frac{\omega_e}{\omega_M}\frac{1}{ka}\frac{M_0 a^3}{\mu_B} \qquad \left(\frac{H}{H_C} \leqslant 1\right), \tag{17}$$

where

$$f\left(\frac{p}{p_k}\right) = \frac{p_k}{p}\frac{\eta_l}{1+\eta_l^2}\left\{\left(1+\left[\left(\frac{p}{p_k}\right)^2-1\right]\frac{1+\eta_l^2}{\eta_l^2}\right)^{1/2}-1\right\}.$$

Substituting (17) in (7) and taking into account the remarks made above concerning the integration interval Δk, we get

$$4\pi\chi'' = f\left(\frac{p}{p_k}\right)\frac{2\pi M_0}{H_C}. \tag{18}$$

The numerical value of the susceptibility (~0.8) agrees well with the experimental data[6-7].

3. DISCUSSION OF RESULTS

The nonlinear detuning and partial-dissipation mechanisms for the establishment of the energy equilibrium in the spin system in the case of NFMR yield for the magnetic susceptibility χ'' a theoretical value which agrees well with experiment. A plot of χ'' against the magnetizing field is shown in Fig. 1. For comparison, the same figure shows a plot of the susceptibility in accordance with the data of Suhl and Gottlieb, which obviously gives values that are too high.

In addition to this main result, it is useful to emphasize two more conclusions of the theory.

First, the results of the NFMR theory require that preference be given to the Bloch model of the magnetization relaxation. This can be readily verified by the following simple reasoning. As is well known, the squares of the total magnetic moment $(\int M dV)^2$ and of its transverse component $(\int M_\perp dV)^2$ are connected with the magnon numbers n_0 and n_k as follows[6]:

$$\left(\int M dV\right)^2 = (M_0 V)^2 - 2\mu M_0 V \sum_{k\neq 0} n_k,$$
$$\left(\int M_\perp dV\right)^2 = 2\mu M_0 V n_0. \tag{19}$$

Here V—volume of the crystal, $\mu = \gamma\hbar$, $\hbar$—Planck's constant, and n_0 has the meaning of the number of magnons with $k < 1/L$ (where L is the largest dimension of the crystal sample). The Landau-Lifshitz model is characterized by a rapid relaxation of the total moment (with a time T_1) and a relatively slow variation of its transverse component (with a time T_2), whereas in the Bloch model the two times are commensurate.

The nonlinear detuning mechanism has no lower limit on the value of the magnon quasimomentum $\hbar k$,

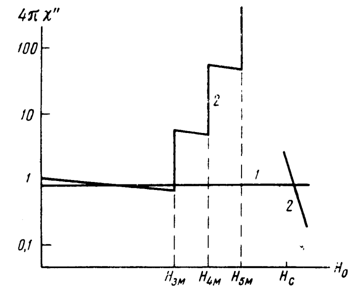

FIG. 1. Plots of the nonlinear magnetic susceptibility $4\pi\chi''$ vs. the magnetizing field H_0. Curve 1 is plotted in accordance with (18) for $\eta_l^2 \ll 1$. The following values of the parameters are used: f = 9300 MHz, $4\pi M_0 = 1.7 \times 10^3$ G, $H_e = 3.5 \times 10^5$ Oe, $k' = k_{max} = 3 \times 10^5$ cm^{-1}. Curve 2 is constructed in accordance with the refined data of the Suhl-Gottlieb theory[1] (it is assumed that $\cos\theta_k = 10^{-1}$ when $H < H_C$). The true calculated susceptibility corresponds to the smallest values obtained from a comparison of the two curves. Experiment yields the value $4\pi\chi'' \cong 0.2$[6].

therefore the relaxation of the transverse component of the macroscopic magnetization and of the total moment are determined by the same magnons, and we therefore arrive at the Bloch model. When $H < H_C$, the macroscopic transverse component of the magnetization of a parametrically excited spin system vanishes. But in this case one can speak of a partial relaxation time T_{2k}, which in the case of the detuning mechanism of equilibrium is commensurate with the time T_1. This result coincides with the conclusion of Bar'yakhtar and Urushadze[8], that the Bloch model is applicable in ferromagnetic resonant absorption of energy, when the condition $\hbar\omega \geq 2\hbar\omega_k$ is satisfied. This condition lifts the limitation from below on the value of the quasimomentum of the magnons taking part in the energy dissipation, just as in the considered case of NFMR in the first zone of parametric excitation.

In the case of the Bloch model, the NFMR magnetization relaxation is unstable in the entire region of its existence[2], and therefore expression (18) for $4\pi\chi''$ gives a maximum value during the NFMR auto-modulation period, and can serve only for a comparative estimate in the sense of Fig. 1. The true value of the nonlinear magnetic susceptibility is obtained when account is taken of the NFMR instability[2].

Second, the theory makes it possible to estimate the magnitude of the gap between the excitation thresholds of NFMR (formula (5)) and the slow magnetization oscillations (auto-modulation of NFMR). From the theory of nonlinear solutions it is known (see, for example,[9]), that the gap is determined by the ratio of the magnitudes of the effects produced by the dissipative and detuning equilibrium mechanisms, i.e., by the parameter η_l introduced above. When $\eta_l \to 0$ both thresholds coalesce, and when $\eta_l \to 1$ the gap increases without limit, so that the nonlinear resonance becomes stable.

The estimate of η_l given above shows that both thresholds are close to each other almost in the entire region of the NFMR, and therefore the observation of slow oscillations of magnetization can be used to indicate the NFMR excitation threshold. However, at certain values of the magnetizing field, the parameter η_l has small singularities which should cause the excita-

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2) Suhl and Gottlieb's value of n_k [1] is obtained in the approximation $\eta_l \gg 1$.

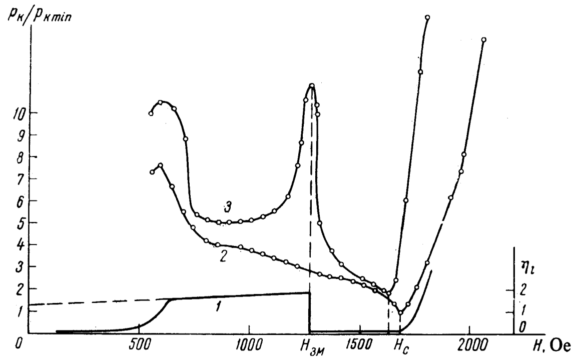

FIG. 2. Plots of the parameter η_l (1) and of the thresholds of parametric excitation of NFMR (2) and auto-modulation of NFMR (3) vs. the magnetic field at f = 9300 MHz (experimental curves 2 and 3 for a spherical single-crystal yttrium-iron garnet oriented along the [001] axis were taken from [10]). The dashed line shows the plot of η_l without allowance for the lowering of the magnetization of the crystal when $H < 4\pi M_0/3$.

tion thresholds to move apart in the experiment.

Figure 2 shows plots of η_l (theory) and of the excitation thresholds of NFMR (known in the literature as a "butterfly" type curve) and of the NFMR auto-modulation (experiment from [10]). We see that the agreement between theory and experiment is not bad.

When $H > 4\pi M_0/3$, the gap between curves 2 and 3 increases smoothly with increasing magnetization field, reaches a maximum at $H = H_{3M}$, and then decreases abruptly. When $H_{3M} < H < H_C$, the curves 2 and 3 almost coalesce, and then when $H > H_C$ the gap again increases. It should be noted that according to the estimate (15) we have $\eta_{3M} > 1$; therefore from the fact that auto-modulation is observed in experiment at $H = H_{3M}$ we can conclude that the theoretical estimate (15) is apparently slightly overvalued.

The extremum on curve 3 explains the small spike on the χ'' curve at field H_{3M}, which is usually observed in the experiments [6,7]. The point is that the auto-modulation decreases the mean value of the susceptibility observed in the experiments, and an increase in the auto-modulation threshold increases the susceptibility somewhat. Such an interpretation of this spike seems to us more natural than the one given in Schlomann's paper [6].

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