Effect of rf modulation on the parallel-pumping threshold in easy-plane antiferromagnets

V. I. Ozhogin, A. Yu. Yakubovskiĭ, A. V. Abryutin, and S. M. Suleĭmanov

I. V. Kurchatov Atomic Energy Institute

(Submitted 22 May 1979)
Zh. Eksp. Teor. Fiz. 77, 2061–2069 (November 1979)

The effect of modulation of the magnetic field (frequency $F = 0.05$ to 2.5 MHz, amplitude $H_m = 0$–2 Oe) on the threshold of parallel microwave pumping ($\omega_p/2\pi = 36.2$ GHz) in the easy-plane antiferromagnets $MnCO_3$ and $CsMnF_3$ is investigated theoretically and experimentally. A mechanism of "indirect" modulation of the "ferromagnon" frequency via participation of an "antiferromagnetic" mode (i.e., of a mode linearly excited by the longitudinal microwave field) is proposed. Good quantitative agreement between theory and experiment is obtained. It is noted that an investigation of the dependence of the pump threshold on the rf modulation frequency serves as an independent method of measuring the paramagnetic-magnon relaxation frequency, and furthermore, does not call for absolute measurements of the alternating-field amplitudes.

PACS numbers: 75.50.Ee, 75.30.Ds, 78.70.Gq

The question of the influence of the high-frequency (RF) modulation of the magnetic field on the threshold of parameteric excitation of magnons was first considered by Suhl,[1] who showed that the threshold should increase in this case because the parametric resonance condition is violated in the mean. A qualitative experimental confirmation of Suhl's theory was obtained by Hartwick, Perressini, and Weiss[2] for yttrium iron garnet (YIG). Zautkin, L'vov, Orel and Starobinets[3] developed a rigorous theory of parametric excitation of magnons in a ferrite in the presence of RF modulation of the constant field and confirmed it quantitatively by experiments on YIG.

The singularities of parametric excitation of magnons in antiferromagnets are due to the presence of two magnon modes, one of which ("quasiferromagnetic") is linearly excited by an alternating magnetic field $\mathbf{h}$ perpendicular to the constant field $\mathbf{H}_0$ while the other ("quasi-antiferromagnetic") is linearly excited at $\mathbf{h} \parallel \mathbf{H}_0$,[4] i.e., by a field having the same polarization that is needed for parametric (nonlinear) excitation of the ferromagnetic mode. Therefore, besides the direct pumping process that takes place in ferrites, in antiferromagnets there is an indirect pumping process of comparable intensity, due this linear excitation of homogeneous oscillations of the "antiferromagnetic" mode and to the nonlinear mode coupling.[5,6]

For parallel microwave pumping of "ferromagnons" in easy-plane antiferromagnets, which are the most convenient from the experimental point of view, the equation for the threshold amplitude, in the case of a pump frequency $\omega_p \ll \omega_{20}$ (ω_{20} is the frequency of the homogeneous resonance of the antiferromagnetic mode), is of the form[5]

$$h_{c0} = \frac{2\eta_{1k}\omega_p}{\gamma^2(2H_0+H_D)}, \tag{1}$$

where ω_p is the pump frequency η_{1k} is the ferromagnon relaxation frequency, H_0 is the constant magnetic field, and H_D is the Dzyaloshinskiĭ field. This simple formula makes it possible to determine the magnon relaxation frequency from parameters measured in experiment. It is obvious, however, that an experimental verification of Eq. (1) itself is possible only if an independent measurement is made of η_{1k}—for example by measuring the transient processes in a system of parametric magnons. It turns out[7] that in experiments of this type the decisive role is the time of loss of phase correlation of magnon pairs in the above-threshold state, which apparently differs from the magnon lifetime determined from the parallel-pumping threshold.

It follows from the results of Ref. 3 that a study of the effect of RF modulation of the constant field on the parallel-pumping threshold also yields information on the relaxation parameter of the excited magnons. We have therefore undertaken an analogous investigation for easy-plane antiferromagnets (AF), paying particular attention to the participation of the antiferromagnetic mode in the formation of the modulation mechanism.

INDIRECT MODULATION MECHANISM

We consider an AF with easy-plane (EP) anisotropy, placed in a field $\mathbf{H} \parallel$ LP at a temperature $T \ll T_N$. Let $H = H_0 + h\cos\omega_p t + H_m \cos\Omega t$, where h is the microwave pump amplitude and H_m is the amplitude of the RF modulation ($\Omega \ll \omega_p$, so that the phase shift between the pump and the modulation can be disregarded). Starting from Eq. (A.1) of the preceding paper[8] for the energy of the electron-spin system and diagonalizing it with the aid of the UV transformation [Eqs. (5a) and (A.1) of Ref. 8], we represent the Hamiltonian of the problem in terms of the normal amplitudes $c_{1\mathbf{k}}, c^*_{1\mathbf{k}}$ and $c_{2\mathbf{k}}, c^*_{2\mathbf{k}}$ of the ferro- and antiferromagnons:

$$\begin{aligned} \frac{1}{\hbar}\mathcal{H} = {} & \omega_{20}c_{20}^*c_{20} + \gamma(h\cos\omega_p t + H_m\cos\Omega t)R_0(c_{20}^* + c_{20}) \\ & + \sum_{\mathbf{k}} [\omega_{1\mathbf{k}} + G_{\mathbf{k}}\gamma H_m \cos\Omega t - \Theta_{\mathbf{k}}(c_{20}^* + c_{20})]c_{1\mathbf{k}}^*c_{1\mathbf{k}} \\ & + \Big\{\sum_{\mathbf{k}} [F_{\mathbf{k}}\gamma h\cos\omega_p t - (\Phi_{\mathbf{k}}^*c_{20}^* + \Psi_{\mathbf{k}}c_{20})]c_{1\mathbf{k}}^*c_{1-\mathbf{k}}^* + \text{c.c.}\Big\}. \end{aligned} \tag{2}$$

All the symbols are those used in Ref. 8, except that the index e is omitted, since we shall deal exclusively with the electronic (e) spin subsystem.

The terms containing $c_{2\mathbf{k}}$ and $c^*_{2\mathbf{k}}$ with $\mathbf{k} \neq 0$ can be disregarded at low temperatures. We have left out also all

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the fourth-order terms, which are needed only for the description of the above-threshold state. In addition, we have omitted the terms

$$G_{\mathbf{k}}\gamma h\cos\omega_p t c_{1\mathbf{k}}^* c_{1\mathbf{k}}, \qquad F_{\mathbf{k}}\gamma h \cos\Omega t c_{1\mathbf{k}}^* \overset{*}{c}_{1-\mathbf{k}}.$$

The former were omitted because the very fast (microwave) modulation of the ferromagnon frequency does not influence the threshold, and the latter because the low-frequency pumping ($\Omega < \omega_{10}$) cannot lead to parametric excitation because the energy conservation (time synchronism) condition cannot be satisfied.

At $k \ll a_0^{-1}$, where a_0 is the lattice constant, the coefficients in (2) take the form

$$R_0=(U_{20}-V_{20})(2NS)^{1/2}, \quad F_{\mathbf{k}}=2U_{1\mathbf{k}}V_{1\mathbf{k}}\sin\psi, \quad G_{\mathbf{k}}=2(U_{1\mathbf{k}}^2+V_{1\mathbf{k}}^2)\sin\psi;$$
$$\Phi_{\mathbf{k}}=2\gamma H_0\cos\psi(U_{20}V_{1\mathbf{k}}^2-V_{20}U_{1\mathbf{k}}^2)/(2NS)^{1/2},$$
$$\Psi_{\mathbf{k}}=2\gamma H_0\cos\psi(U_{20}U_{1\mathbf{k}}^2-V_{20}V_{1\mathbf{k}}^2)/(2NS)^{1/2},$$
$$\Theta_{\mathbf{k}}=4\gamma H_0\cos\psi(U_{20}-V_{20})U_{1\mathbf{k}}V_{1\mathbf{k}}/(2NS)^{1/2},$$

where ψ is the equilibirum sublattice cant angle, defined by the equation

$$H_E\sin 2\psi=H_D\cos 2\psi+H_0\cos\psi.$$

The terms with $F_{\mathbf{k}}$ and $G_{\mathbf{k}}$ describe the "direct" pump and modulation processes, respectively, and stem from the Zeeman term in the initial expression for the energy. On the other hand the terms with $\Phi_{\mathbf{k}}$, $\Psi_{\mathbf{k}}$, and $\Theta_{\mathbf{k}}$ are due to the nonlinearity of the symmetric and antisymmetric exchange interactions between the sublattices and their contribution to the pump and to the modulation is "indirect" and typical only of AF: the term with R_0 makes possible linear excitation, at the frequencies ω and Ω, of the homogeneous antiferromagnetic mode up to amplitudes c_{20} proportional to h and H_m, and these, owing to the three-particle terms with the coefficients $\Phi_{\mathbf{k}}$, $\Psi_{\mathbf{k}}$, and $\Theta_{\mathbf{k}}$ renormalize both the pump (see Refs. 5 and 6) and the modulation.

Under the most frequently encountered experimental conditions(Ω, ω_p, $\gamma H_0 \ll \omega_{20} \ll \omega_E$) the expressions for these coefficients are greatly simplified:

$$2F_{\mathbf{k}}\approx G_{\mathbf{k}}\approx\gamma(H_0+H_D)/2\omega_{1\mathbf{k}},$$
$$2R_0\Phi_{\mathbf{k}}\approx 2R_0\Psi_{\mathbf{k}}\approx R_0\Theta_{\mathbf{k}}\approx\gamma H_0\omega_{20}/4\omega_{1\mathbf{k}}.$$

Then the equations of motion stemming from the Hamiltonian (2) yield for the amplitude of the homogeneous ($k=0$) antiferromagnetic mode

$$c_{20}\approx-\gamma h R_0 e^{i\omega_p t}/2\omega_{20}-\gamma H_m R_0 e^{i\Omega t}/2\omega_{20}+\text{c.c.} \tag{3}$$

Substituting (3) in (2) we obtain the renormalized Hamiltonian

$$\frac{1}{\hbar}\tilde{\mathcal{H}}=\sum_{\mathbf{k}}(\omega_{1\mathbf{k}}+2\tilde{U}_{\mathbf{k}}H_m\cos\Omega t)c_{1\mathbf{k}}^*c_{1\mathbf{k}}$$
$$+\Big[\sum_{\mathbf{k}}\tilde{V}_{\mathbf{k}}h\cos\omega_p t c_{1\mathbf{k}}^*\overset{*}{c}_{1-\mathbf{k}}+\text{c.c.}\Big], \tag{4}$$
$$\tilde{U}_{\mathbf{k}}\approx\tilde{V}_{\mathbf{k}}\approx\gamma^2(2H_0+H_D)/4\omega_{1\mathbf{k}}. \tag{4a}$$

The reasoning of Ref. 3 is now fully applicable; in particular, for not too large $H_m < H_{m\,\text{cr}}$ and not too small $\Omega > 2\eta_{1\mathbf{k}}$ we easily obtain an expression that describes the influence of the RF modulation on the threshold h_c of the parametric excitation of ferromagnons with $\omega_{1\mathbf{k}} = \omega_p/2$:

$$\frac{h_c}{h_{c0}}-1=\frac{\gamma^4 H_m^2(2H_0+H_D)^2}{\omega_p^2(\Omega^2+4\eta_{1\mathbf{k}}^2)}, \tag{5}$$

where h_{c0} is the threshold pump amplitude in the absence of modulation, see Eq. (1).

As already noted, expression (1) for h_{c0} cannot be verified in experiment, since it contains a material characteristic $\eta_{1\mathbf{k}}$ that is not directly measurable. On the other hand expression (5), which contains $\eta_{1\mathbf{k}}$ in combination with the experimentally set modulation frequency Ω, lends itself fully to direct verification and furthermore does not require the usually difficult absolute measurement of h_{c0}. But since the expression for h_{c0} and h_c were obtained within the framework of the same computation scheme, an experimental verification of (5) can be regarded also as a check on the validity of Eq. (1), the latter being customarily used for the measurement of magnon relaxation frequency in AF by the parallel-pumping method.

EXPERIMENT AND DISCUSSION OF RESULTS

The measurements were made by the standard procedure[8] in a pulsed regime (the duration of the synchronized pump and modulation pulses was 200 μsec) at a pump frequency $\omega_p/2\pi = 36.2$ GHz. The investigated sample was placed in a TE_{012} resonator and subject to three parallel magnetic fields in the basal (easy) plane of the crystal: the constant field H_0, the microwave pump $h\cos\omega_p t$, and the RF modulation $H_m\cos\Omega t$. The modulation field with amplitude $H_m = 0-2$ Oe and frequency $\Omega/2\pi \equiv F = 0.05-2.5$ MHz was produced by a single-turn coil located on the resonator axis and containing the sample (a plate measuring ~$3\times 2\times 0.5$ mm). The presence of the modulation coil decreased the Q of the resonator by one-half (to $Q = 1500$). All the measurements were made at $T = 1.7$ K. To assess the role of the Dzyaloshinskiĭ interaction we investigated the single crystals $MnCO_3$($H_D = 4.4$ kOe) and $CsMnF_3$($H_D = 0$).

Figure 1 shows the relative increase of the parallel-pump threshold in $MnCO_3$ as a function of the RF modulation frequency F at different H_m. These functions are plotted in Fig. 2 in coordinates for which Eq. (5) predicts a straight line:

$$(h_c/h_{c0}-1)^{-1}=\alpha F^2+\beta; \tag{6}$$

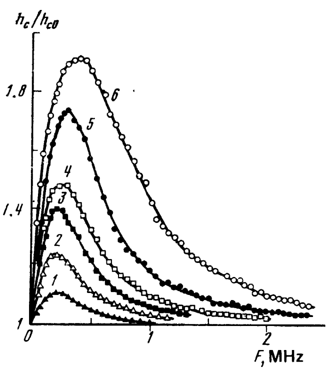

FIG. 1. Relative increase of the threshold of parallel pumping in $MnCO_3$ as a function of the RF modulation frequency: $H_0 = 0.8$ kOe; curves: 1—$H_m = 0.1$ Oe, 2—0.15 Oe, 3—0.2 Oe, 4—0.25 Oe, 5—0.35 Oe and 6—0.5 Oe.

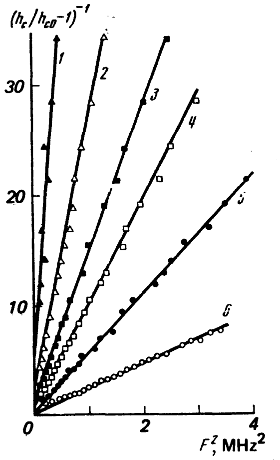

FIG. 2. Plots of the relative increase of the parallel-pumping threshold in $MnCO_3$ against the RF modulation frequency in coordinates in which the theory [Eq. (5)] predicts straight lines; $H_0=0.8$ kOe, $T=1.7$ K; curves: 1—$H_m=0.1$ Oe, 2—0.15 Oe, 3—0.2 Oe, 4—0.25 Oe, 5—0.35 Oe and 6—0.5 Oe.

here

$$\alpha=(2\pi/U)^2/4H_m^2,\quad \beta=\alpha\Delta\nu_k^2\quad (\Delta\nu_k=\eta_{1k}/\pi); \tag{6a}$$

$$U=\gamma^2(2H_0+H_D)/2\omega_p. \tag{7}$$

The experimental points are seen to fit straight lines well. Discrepancies are observed only at low frequencies, where the theory developed by Zautkin *et al.*[3] does not hold (in Fig. 1 this region corresponds to the steep slopes of the curves at low frequencies). The deviations from the straight lines are due to the fact that the theory assumes modulation of the natural frequency of the magnons ω_{1k} without a shift of the magnon packet in k space. It is obvious that this condition is not satisfied at sufficiently low frequencies: the packet follows the k variation, which is determined by the parametric-resonance condition $\omega_{1k}(H)=\omega_p/2$, more accurately the lower the modulation frequency. By the same token, the effect of modulation on the threshold of the parallel pumping is decreased.

Figure 3 shows the dependence of the relative increase of the threshold of the parallel pumping in $MnCO_3$ on the RF modulation frequency. The initial quadratic growth saturates gradually. A similar explanation of this behavior of the curves can be found also in Ref. 3. We are interested in the region of H_m and F where a quadratic growth of the threshold is observed with increasing H_m and F^{-1} and it can be assumed that the minimal threshold is reached for magnons with $\omega_{1k}=\omega_p/2$. All the subsequent measurements were made just in this region, where, in particular, we can disregard the dependence of the coefficient $\tilde{U}_k$ on k and therefore put $\tilde{U}_k=U$ [see (7)].

Equations (6a) predict a linear dependence of the quantities α and β on H_m^{-2}. Figure 4 shows the dependence of the slopes of the straight lines of Fig. 2 on H_m^{-2}. The slope of the obtained straight line is 0.54 ± 0.1 Oe^2/MHz^2, in good agreement with the value 0.59 Oe^2/MHz^2 calculated from Eqs. (6a) and (7). The plot of $\beta(H_m^{-2})$ is also a straight line, from the slope of which

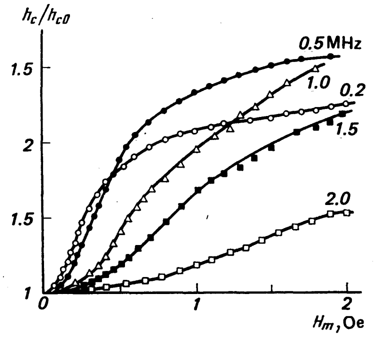

FIG. 3. The relative increase of the threshold of parallel pumping in $MnCO_3$ vs. the RF modulation amplitude; $H_0=0.8$ kOe, $T=1.7$ K, and the frequency F is marked on the curves.

we can determine the relaxation frequency of the parametric magnons for $MnCO_3$ in a field $H_0=0.8$ kOe at $T=1.7$ K:

$$\eta_{1k}=\pi\Delta\nu_k=(1\pm0,1)\cdot10^6\ \mathrm{sec}^{-1}.$$

Figure 5 shows plots of the relative increase of the threshold of the parallel pumping in $MnCO_3$ on the square of the modulation frequency at fixed $H_m=0.25$ Oe and different values of H_0. Analyzing the dependence of the slopes of these straight lines on the parameter $(2\pi/U)^2$, we can verify the assumption that an indirect contribution is made to modulation of the ferromagnons. In accord with (6a), this plot is a straight line with a slope 3.75 Oe^{-2} that agrees well with the calculated value $1/4H_m^2=4\pm0.8$ Oe^{-2}. Figures 7 and 8 show the analogous plots for $CsMnF_3$. The slope 0.9 Oe^{-2} of the line in Fig. 8 again agrees well with the calculated value $1/4H_m^2=1\pm0.2$ Oe^{-2}.

The fact that the plots in Figs. 6 and 8 are straight lines having a slope that agrees well with the calculation in which it is assumed that $U=\gamma^2(2H_0+H_D)/2\omega_p$ attests to the validity of the developed theoretical premises concerning the mechanism of the modulation of the frequency of the paramagnetic magnons: the contribution made to the quantity $U\cdot2\omega_p/\gamma^2$ by the direct modulation process amounts to H_0+H_D, while the contribution from the indirect process is H_0. This can be regarded as a quantitative confirmation of the existence of the indirect mechanism of modulation with participation of the upper magnon mode. Since the indirect modulation and the indirect parallel pumping have identical mechanisms, the verification of Eq. (5) can be regarded also as a confirmation of the validity of Eq. (1) for the

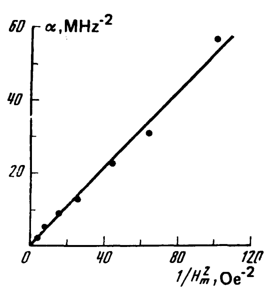

FIG. 4. Plot of the coefficient α [see Eq. (6a)] against H_m^{-2}, obtained from the results of Fig. 2.

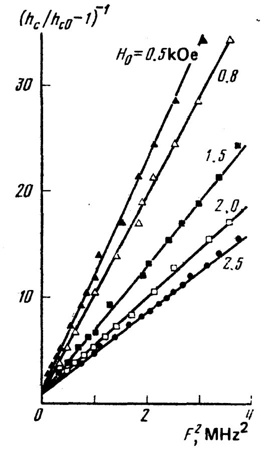

FIG. 5. Relative increase of the threshold of parallel pumping in $MnCO_3$ as a function of the RF modulation frequency; $H_m = 0.25$ Oe, $T = 1.7$ K; the values of H_0 are marked on the curves.

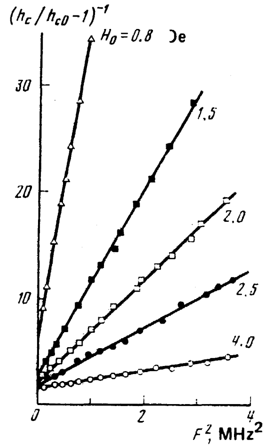

FIG. 7. Relative increase of the parallel pumping threshold in $CsMnF_3$ as a function of the RF modulation frequency; $H_m = 0.5$ Oe, $T = 1.7$ K; the values of H_0 are marked on the curves.

threshold of parallel microwave pumping in easy-plane AF.

The stage-by-stage comparison of experiment with theory, made in Figs. 4–8, confirms the validity of Eq. (5). Our main experimental result can therefore be represented quite illustratively as the following. Let W be a combination of the parameters measured directly in the experiment:

$$W = \frac{(h_c/h_{c0}-1)^{1/2}\,\Omega\omega_p}{\gamma^2 H_m}. \tag{8}$$

It follows from (5) that at $\Omega^2 \gg 4\eta_{1k}^2$ the following equality should hold:

$$W = 2H_0 + H_D. \tag{9}$$

The solid lines in Fig. 9 are plots of Eq. (9), while the points were calculated with the aid of (8) and correspond to the experimental data averaged over many measurements at various $H_m < 1$ Oe and $F > 1.2$ MHz. It is seen that the experiment agrees well with the calculation. We note by way of a curiosity that Fig. 9 demonstrates also an independent method of measuring H_D.

MODULATION METHOD OF MEASURING THE MAGNON RELAXATION FREQUENCY

Analyzing the slopes of the lines in Figs. 5 and 7, we determined the coefficient U and obtained useful information concerning the magnon frequency modulation mechanism. We consider now the intercepts of these lines with the ordinate axis, which we shall designate by β. It follows from (6) and (7) that the magnon relaxation frequency is

$$\eta_{1k} \equiv \pi\Delta\nu_k = UH_m\beta^{1/2} = \frac{\gamma^2(2H_0+H_D)H_m\beta^{1/2}}{2\omega_p}. \tag{10}$$

Consequently the method of RF modulation of the magnetic field makes it possible to measure also the relaxation frequency of the parametric magnons. To this end, as follows from (6), it is sufficient to measure the frequency dependence of the relative increase of the threshold at fixed values of H_0, H_m, and ω_p and calculate $\Delta\nu_k = (\beta/\alpha)^{1/2}$.

The magnon relaxation frequency was previously obtained principally by measuring the absolute value of h_{c0} of the threshold amplitude of the parallel pumping. This calls for an exact calculation of the distribution of the microwave magnetic field in the resonator, as well

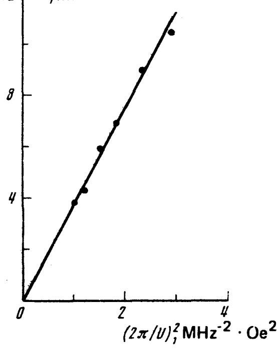

FIG. 6. Plot of the coefficient α against $(2\pi/U)^2$ based on the results of Fig. 5.

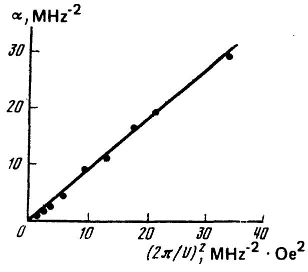

FIG. 8. Plots of the coefficient α against $(2\pi/U)^2$ based on the results of Fig. 7.

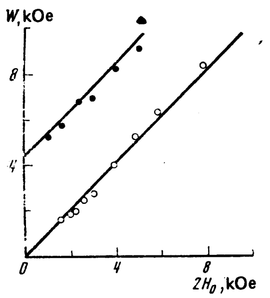

FIG. 9. Dependence of the function W [see Eq. (8)] on $2H_0$. Solid lines—calculation by Eq. (9), points—averaged results of measurements made at different $F > 1.2$ MHz and $H_m < 1$ Oe; $T = 1.7$ K, $\omega_p/2\pi = 36.2$ GHz.

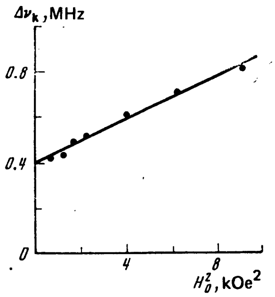

FIG. 10. Relaxation parameter $\Delta\nu_k \equiv \eta_{1k}/\pi$ of parmetric magnons of $CsMnF_3$ measured by the RF modulation method at $T = 1.7$ K as a function of H_0^2; $\omega_p/2\pi = 36.2$ GHz.

as measurements of the microwave power and of the Q and coupling coefficient of the resonator. The use of the modulation procedure greatly simplifies the measurement procedure, and under favorable conditions ($H_D = 0$, see below) the accuracy of the measurement of the magnon relaxation frequency is within several per cent.

We were unable to measure the dependence of the magnon relaxation frequency on H_0 for $MnCO_3$ because the presence of $H_D = 4$ kOe masks the variation of the combination $2H_0 + H_D$ at small H_0 and the intercepts β at various H differ little from one another. In $CsMnF_3$ we have $H_D = 0$, and change of H_0 leads to a noticeable change of β (see Fig. 7). For $CsMnF_3$ we can therefore obtain formally the relaxation frequencies $\eta_1[\mathbf{k}(H_0)]$ as functions of H_0^2 (Fig. 10). This quadratic dependence agrees with the results of earlier measurements of this quantity,[9] in which Eq. (1) was used.

Kotyuzhanskiĭ and Prozorova[9] have proposed to regard $\Delta\nu_k$ as a sum of two terms: the first, $\Delta\nu_{intr}(H_0, T)$ is due to collisions between the quasiparticles, and the second $\Delta\nu_{res}(\mathbf{k})$ is determined by the dimensions and quality of the sample, and is measured as the limit of $\Delta\nu_k$ as $H_0 \to 0$. A comparison of the absolute values of $\Delta\nu_k$ obtained for $CsMnF_3$ in Ref. 9 and in the present study (Fig. 10) reveals a noticeable discrepancy (by an approximate factor of 3) in both $\Delta\nu_{res}$ and $\Delta\nu_{intr}$. The relaxation parameter measured by us in $MnCO_3$, $\Delta\nu_{res} = 0.32$ MHz, is also approximately 3 times larger than the results of measurements based on the parallel-pumping threshold,[10] made on samples from the same source as used by us. This discrepancy possibly calls for a more careful examination of the question of the identity of the relaxation parameters that enter in expressions (1) and (5).

We are deeply grateful to I. K. Kikoin for interest in this work and L. A. Prozorova for helpful discussions.

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Translated by J. G. Adashko