

# Modulation-method investigation of the steady state of parametric spin waves in antiferromagnets

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The response of parametric electron and nuclear spin waves in the antiferromagnets  $\text{MnCO}_3$  and  $\text{CsMnF}_3$  to a weak modulation of a static magnetic field was investigated. A calculation was made of the modulation response allowing for the phase mechanism limiting the amplitude of parametric waves and for their positive nonlinear damping. An analysis of the experimental results carried out within the framework of this model made it possible to determine the characteristics of the steady state of parametric nuclear magnons: the coefficient  $S_k$  representing a nonlinear magnon interaction and the parameter  $\kappa$  representing the relative contributions of positive nonlinear damping and of the phase mechanism to limiting the number of parametric magnons. An anomaly was observed in the behavior of the modulation response of parametric nuclear magnons in  $\text{CsMnF}_3$ , which was manifested by a strong weakening of the response in a narrow range of supercriticalities and modulation frequencies. The same crystal exhibited a giant hexagonal anisotropy of the modulation response and a nonlinear dynamic susceptibility of nuclear magnons which could indicate their nonisotropic distribution in the  $k$  space.

## INTRODUCTION

Experiments on parametric pumping of spin waves by a microwave field  $h \cos \omega_p t$  ( $\mathbf{h} \parallel \mathbf{H}_0$ , where  $\mathbf{H}_0$  is a constant magnetic field) are important in the investigation of the properties of magnetically ordered materials. The process of parametric excitation is characterized by a threshold or critical amplitude  $h_c$ , beginning from which a spin system exhibits an instability against a decay of a pump quantum into a pair of electron (*ee* process) or nuclear (*nn* process) magnons of half the frequency with oppositely directed wave vectors ( $\omega_p = \omega_{\mathbf{k}} + \omega_{-\mathbf{k}}$ ). The value of  $h_c$  is used to determine the rate of relaxation of excited modes, and an above-threshold ( $h > h_c$ ) state of parametric waves is a convenient object for the investigation and simulation of the processes occurring in a nonlinear many-particle medium. We shall report an investigation of an above-threshold state of a parametric system of electron nuclear spin waves in antiferromagnets  $\text{MnCO}_3$  and  $\text{CsMnF}_3$ .

Two mechanisms are known to limit the amplitudes of excited spin waves beyond the parametric resonance threshold. The first is due to an increase in the dissipation of modes on increase of their intensity and is called the nonlinear damping mechanism.<sup>1</sup> The second is due to a mismatch between parametric oscillations of the medium and the pump, and is called the phase mechanism.<sup>2</sup>

In a real parametric system these two mechanisms coexist, but so far the experimental results have been analyzed in those cases when one of them can be ignored. According to the theory of Ref. 2 (*S* theory), an excited system is characterized by the number of parametric magnon pairs  $N_k$  and by the phase  $\theta_k$  of their mismatch relative to the pump ( $\theta_k = \pi/2 - \psi_k$ , where  $\psi_k$  is the temporal phase of a pair). It follows from the *S* theory that in the case of antiferromagnets for which it is possible to ignore the dipole anisotropy of the amplitudes of the magnon-magnon interactions (see Ref. 3), a spherically symmetric (in respect of  $\mathbf{k}$ ) stable steady state of the parametric system with specific values of

$N_k^{(0)}$  and  $\theta_k^{(0)}$  is realized above the excitation threshold of spin waves. The properties of this state had been investigated experimentally on the basis of the nonlinear dynamic magnetic susceptibility  $\chi = \chi' + i\chi''$  of the spin system (*ee* in Refs. 4 and 5; *nn* in Ref. 6), by the method of transient processes (*ee* in Ref. 7), on the basis of radiation emitted by a parametric system (*ee* in Refs. 8 and 9), and by a modulation method (*nn* in Ref. 10). The experimental results obtained in Refs. 4, 6, 7, 9, and 10 for  $\text{MnCO}_3$  and  $\text{CsMnF}_3$  crystals were in satisfactory agreement with the main conclusions of the *S* theory, whereas the experimental data obtained in Refs. 5 and 8 for  $\text{FeBO}_3$  agreed with the model of nonlinear damping of spin waves.

It would be of interest to carry out a detailed investigation of the properties of a steady state of parametric spin waves allowing simultaneously for two limitation mechanisms. Theoretically this problem can be solved by including the nonlinear damping of spin waves into the equations of the *S* theory. It is natural to assume that this procedure should bring the range of validity of the theory closer to the experimental conditions.

A modulation method for the investigation of the properties of a steady state of spin waves was proposed in Ref. 10: it involves a study of the response of a system of parametric magnons to a weak harmonic perturbation  $H_m \cos \omega_m t$  of an external magnetic field  $\mathbf{H}_0$  ( $\mathbf{H}_m \parallel \mathbf{H}_0$ ,  $\omega_m \ll \omega_p$ ). This perturbation modulates the spectrum of spin waves, which in turn results in amplitude modulation of the microwave power absorbed by a sample. At low values of  $H_m$  the amplitude  $\Delta P$  of the oscillations of the absorbed microwave power is

$$\Delta P = \alpha_m H_m, \quad (1)$$

where  $\alpha_m$  is the "modulation response" (see Ref. 11) dependent on the parameters of the steady state of the excited system.

We shall report experimental and theoretical investigations of a steady state of a parametric system carried out on

the basis of a model which allows both for the phase mechanism limiting the amplitude of the excited spin waves and for the nonlinear damping of these waves.

### CALCULATION OF THE MODULATION RESPONSE

We shall calculate the modulation response  $\alpha_m$  using a phenomenological  $S$  theory<sup>2</sup> allowing for the nonlinear damping of waves. Oscillations of the power absorbed in a sample from two alternating magnetic fields are described by the expression

$$\delta P(t) = 2\omega_p \mu_{eff} \hbar \delta [N_k(t) \cos \theta_k(t)] + 2\omega_m \mu_{eff} H_m N_k \sin \omega_m t, \quad (2)$$

where  $\mu_{eff} = \hbar \partial \omega_k / \partial H$  is the effective magneton of the excited magnons. In the calculation of  $\Delta P$  only the contributions<sup>1)</sup> linear in  $H_m$  and oscillating at a frequency  $\omega_m$  need be retained in the first and second terms of Eq. (2). The quantities  $N_k$  and  $\theta_k$  obey the equations<sup>2,10</sup>

$$\frac{1}{2} \frac{d}{dt} \theta_k + \gamma_k b \sin \theta_k = \Delta_k(t) - \frac{S_k N_k}{\mathcal{N}}, \quad (3a)$$

$$\frac{1}{2} \frac{d}{dt} N_k = N_k \gamma_k \left( b \cos \theta_k - 1 - \frac{\tilde{\gamma}_k}{\gamma_k} \right), \quad (3b)$$

where

$$\Delta_k(t) = \omega_p / 2 - \tilde{\omega}_k(t),$$

$$\tilde{\omega}_k(t) = \omega_k + 2T_k N_k / \mathcal{N} + (\partial \omega_k / \partial H) H_m \cos \omega_m t$$

is the renormalized frequency of spin waves which allows for the modulation field;  $S_k$  and  $T_k$  are the coefficients of the nonlinear interaction of magnons<sup>2</sup> (in the isotropic case we have  $S_k = T_k$ );  $b \equiv \hbar / h_c$ ;  $\mathcal{N}$  is the number of magnetic cells in a sample;  $\gamma_k$  is the rate of relaxation of spin waves in the absence of pumping (linear relaxation);  $\tilde{\gamma}_k(N_k)$  is a correction to the rate of relaxation of spin waves which appears above the threshold of the parametric excitation of magnons (nonlinear damping).

The steady-state values of  $N_k^{(0)}$  and  $\theta_k^{(0)}$  are found from the system of equations (3) where the time derivatives and  $\Delta_k$  are assumed to vanish.<sup>2)</sup> Introducing

$$N_k = N_k^{(0)} + \delta N_k, \quad \theta_k = \theta_k^{(0)} + \delta \theta_k,$$

we solve the system (3) in the approximation linear in  $H_m$ . We retain the first term of the expansion of  $\tilde{\gamma}_k$  in terms of  $N_k$ :

$$\tilde{\gamma}_k = \gamma_k \eta N_k / \mathcal{N}, \quad (4)$$

where  $\eta$  is the nonlinear damping coefficient. In this case we have

$$N_k^{(0)} = \mathcal{N} \frac{\gamma_k}{|S_k|} \frac{[(b^2 - 1)(1 + \kappa^2) + \kappa^2]^{1/2} - \kappa}{(1 + \kappa^2)}, \quad (5)$$

where  $\kappa \equiv \gamma_k \eta / |S_k|$  is a parameter representing the ratio of the contributions of the positive nonlinear damping and phase mechanisms limiting the number of parametric magnons.

It should be pointed out that in the case of modulation of the spectrum of spin waves the parametric resonance condition  $\omega_p / 2 = \tilde{\omega}_k$  (valid when  $H_m = 0$ ) can be used only at relatively high modulation frequencies when just one mode is excited and the degree of excitation of other modes does

not reach values which could give rise to a comparable response in a time  $t \sim \omega_m^{-1}$ . On the other hand, at low modulation frequencies there is a constant change of excited waves which are in exact resonance with the pump and those which are no longer in resonance. The modes which are no longer in resonance are damped, but as long as their amplitudes are sufficiently high, they still contribute to the overall response of the system. A rigorous solution of the problem of the integrated modulation response of parametric magnons requires information on the evolution of a packet of excited modes in an alternating magnetic field. At present there is no such theory. We shall consider the next simplest "tracking" model in which it is assumed that the parametric system follows changes in the magnetic field. We shall write down the parametric resonance conditions in the form

$$\omega_p / 2 = \langle \tilde{\omega}_k(t) \rangle, \quad (6)$$

where

$$\langle \tilde{\omega}_k(t) \rangle = \int_{-\infty}^t N_k(\tau) \tilde{\omega}_k(\tau) d\tau / \int_{-\infty}^t N_k(\tau) d\tau. \quad (6a)$$

Essentially, Eq. (6) represents the natural assumption that the pump field interacts most strongly with the "center of gravity" of a packet of parametric magnons. In practical calculations it is sufficient to adopt a rough approximation:  $N_k(\tau) \neq 0$  when  $t - t_0 < \tau < t$  and  $N_k = 0$  when  $\tau < t - t_0$ , so that

$$\langle \tilde{\omega}_k(t) \rangle \approx \frac{1}{t_0} \int_{t-t_0}^t \tilde{\omega}_k(\tau) d\tau, \quad (6b)$$

where  $t_0$  is the characteristic correlation time between excited modes in a parametric system under modulation conditions. We shall assume that  $t_0$  is practically independent of  $H_m$  and  $\omega_m$ .

The condition (6) reflects the fact that at low modulation frequencies the parametric system begins to "track" or follow the changes in the magnetic field and thus "screens" the influence of modulation on the value  $\Delta_k(t)$ . Consequently, on the right-hand side of Eq. (3a) we have to replace  $\Delta_k(t)$  with

$$\Delta_k(t) - \langle \Delta_k(t) \rangle = - \frac{\partial \omega_k}{\partial H} H_m (\cos \omega_m t - \langle \cos \omega_m t \rangle) - \frac{2T_k}{\mathcal{N}} [N_k(t) - \langle N_k(t) \rangle]. \quad (7)$$

Obviously, the screening effect becomes important at frequencies  $\omega_m \lesssim t_0^{-1}$ , whereas in the range  $\omega_m \gg t_0^{-1}$  the screening is lost completely and the parametric resonance condition of Eq. (6) reduces to its usual form.

Equations (2) and (3) yield the following expression for the oscillations of the absorbed microwave power:

$$\delta P(t) = \alpha_m H_m \sin(\omega_m t + \varphi), \quad (8)$$

where

$$\alpha_m = [\alpha_1^2 + 2\alpha_2 \alpha_1 \sin \varphi + \alpha_2^2]^{1/2},$$

$$\alpha_1 = 2\mu_{eff} \omega_p \frac{\mathcal{N} \gamma_k}{|S_k|} \left\{ [(1 + 2\kappa \kappa_0)^2 + \Omega^2] \frac{A^2 + B^2}{x^2 + y^2} \right\}^{1/2};$$

$$\alpha_2 = 2\mu_{eff} \omega_m N_k^{(0)}. \quad (9)$$

The following notation is used above:

$$\begin{aligned}
\operatorname{tg} \bar{\varphi} &= \alpha_1 \cos \varphi / (\alpha_2 + \alpha_1 \sin \varphi), \\
\operatorname{tg} \varphi &= (By_1 + Ax_1) / (Bx_1 - Ay_1), \\
A &= 1 - \sin \omega_m t_0 / \omega_m t_0, \quad B = (1 - \cos \omega_m t_0) / \omega_m t_0, \\
x &= (\Omega / n_0^2) (1 + 2\kappa n_0) + 2\xi B, \quad \xi = T_k / S_k, \\
y &= (\Omega / n_0)^2 - (1 + \kappa n_0) \kappa / n_0 - 2\xi A - 1, \\
x_1 &= x(1 + 2\kappa n_0) + \Omega y, \quad \Omega = \omega_m / 2\gamma_k, \\
y_1 &= y(1 + 2\kappa n_0) - \Omega x, \quad n_0 = (|S_k| / \gamma_k) N_k^{(0)} / \mathcal{N}^0. \quad (10)
\end{aligned}$$

We shall now introduce the asymptotes of  $\alpha_m$ :

$$\begin{aligned}
\text{a) } \alpha_m &\propto \omega_m \text{ for } \omega_m \rightarrow 0, \\
\text{b) } \alpha_m &\propto \omega_m^{-1} \text{ for } 2(\gamma_k + \tilde{\gamma}_k) \ll \omega_m \ll \omega_m^*, \\
\text{c) } \alpha_m &\propto \omega_m \text{ for } \omega_m \gg \omega_m^*. \quad (11)
\end{aligned}$$

where  $\omega_m^*$  are found from the condition  $\alpha_1 = \alpha_2$ , which gives

$$\omega_m^* = [2\omega_p |S_k| N_k^{(0)} / \mathcal{N}^0]^{1/2}.$$

In the case of typical values of the parameters of nuclear spin waves in antiferromagnets, we have  $\omega_m^* / 2\pi \sim 10^7$  Hz, so that in the range of frequencies  $\omega_m / 2\pi \lesssim 10^6$  Hz of interest to us the coefficient  $\alpha_2$  in Eq. (9) can be ignored.

We shall now consider the modulation response of  $\alpha_m$  as a function of three parameters:  $\kappa$ ,  $S_k$ , and  $\tau_0^{-1} = 2\gamma_k t_0^{-1}$  (we recall that the quantity  $\gamma_k$  is calculated from the excitation threshold of a parametric resonance). These parameters are obtained from an analysis of the frequency dependences of  $\alpha_m$  (Ref. 11), which have a characteristic maximum. The width and the position of this maximum depend mainly on  $\kappa$ , the nature of the fall of  $\alpha_m$  at low frequencies allows us to find  $\tau_0^{-1}$ , and the absolute magnitude of the modulation response is used to calculate the coefficient  $S_k$ . In the description of the experimental results we shall use the following frequency scale:

$$\nu_j \equiv \omega_j / 2\pi \quad (j=p, m), \quad \Gamma_k \equiv \gamma_k / 2\pi.$$

## EXPERIMENTAL METHOD

Spin waves were excited in antiferromagnetic crystals  $\text{MnCO}_3$  and  $\text{CsMnF}_3$  by the method of parallel pumping at a frequency  $\nu_p = 22.7$  GHz (*ee* process) and at frequencies  $\nu_p = 0.7$ – $1.0$  GHz (*nn* process). The excitation method was described in detail in Refs. 7 and 13. In addition to a constant magnetic field  $H_0$  and a microwave pump field  $h \cos(2\pi\nu_p t)$  a sample was subjected to a parallel field  $H_m \cos(2\pi\nu_m t)$  of frequency which was varied in the range  $\nu_m = 0$ – $600$  kHz (in the case of nuclear spin waves) and  $\nu_m = 0$ – $5$  MHz (in the case of electron spin waves). The amplitude of the modulation field  $H_m$  was selected so that the influence of this field on the threshold  $h_c$  could be ignored.

The modulation response  $\alpha_m$  was deduced from the depth of amplitude modulation of the microwave signal at the output of a resonator containing a sample; this was done using a spectrum analyzer or a selective microvoltmeter. The frequency characteristic of the receiving channel was determined in a preliminary experiment and the channel was calibrated when the absolute values of  $\alpha_m$  were required. The  $\alpha_m(\nu_m)$  dependences were usually determined at fixed values of  $H_m$ ,  $h/h_c$ ,  $H_0$ , and  $T$  and then the results obtained were analyzed in order to determine the parameters of a steady state of the investigated crystal.

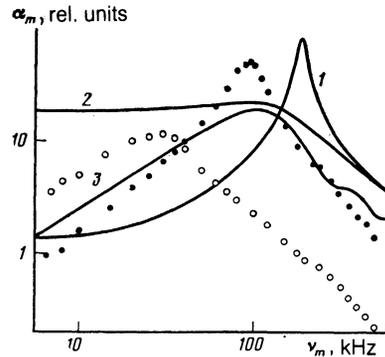


FIG. 1. Frequency dependences of the modulation response of nuclear spin waves in  $\text{MnCO}_3$  obtained for  $\nu_p = 1016$  MHz and  $H_0 = 0.6$  kOe at  $T = 2$  K and different values of supercriticality  $h/h_c$ : 1.64 (O) and 4.73 (●). The continuous curves represent the results of calculations on the assumption that  $2\Gamma_k = 23$  kHz: 1), 2) calculated using Eq. (9) for  $\kappa = 0$  and  $\kappa = 1.1$ , respectively ( $\tau_0^{-1} = 0$ ); 3)  $\kappa = 1.1$  and  $\tau_0^{-1} = 1.5$ .

## STEADY STATE OF A SYSTEM OF PARAMETRIC NUCLEAR SPIN WAVES

In this section we shall give the results of our investigation of the modulation response of parametric nuclear spin waves. Typical experimental  $\alpha_m(\nu_m)$  dependences obtained for  $\text{MnCO}_3$  at two values of the supercriticality (excess about the threshold) are plotted in Fig. 1. These dependences were found to have a characteristic maximum whose position shifted on increase in the supercriticality  $h/h_c$  toward higher frequencies (Fig. 2) and the value of  $\alpha_m$  rose monotonically on increase in the supercriticality until the frequency of the peak  $\nu_m^{\max}$  exceeded  $\nu_m$  (Fig. 3); it should be noted that the results plotted in Figs. 1–3 were obtained under the same experimental conditions. It was easiest to analyze the experimental results on the basis of Eq. (9) at high modulation frequencies when the phenomenological parameter  $\tau_0^{-1}$  had practically no influence on the value of  $\alpha_m$  and the nature of the curves depended only on  $\kappa$ . Then, the error in the determination of  $\kappa$  depended primarily on the linear rate  $\Gamma_k$  of the relaxation of nuclear spin waves calculated from the excitation threshold. For example, the best description of the data in Figs. 2 and 3 was obtained on the assump-

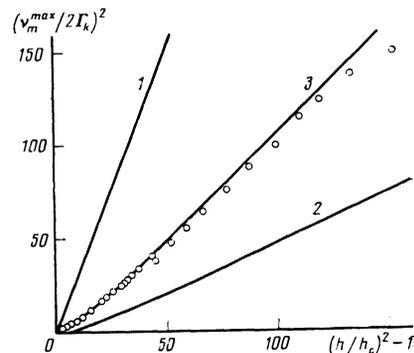


FIG. 2. Dependence of the position of the modulation response maximum on the supercriticality in the case of  $\text{MnCO}_3$ . The best description (curve 3) corresponds to  $\kappa = 1.1$ . Curve 1 is plotted for  $\kappa = 0$  and curve 2 for  $\kappa = 1.5$ ;  $\tau_0^{-1} = 0$ .

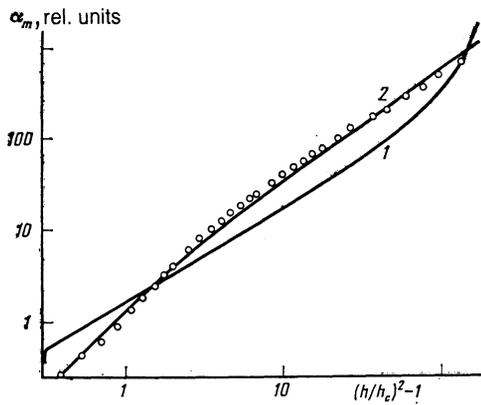


FIG. 3. Dependence of the modulation response of  $\text{MnCO}_3$  at  $\nu_m = 570$  kHz on the supercriticality. Curve 1 corresponds to  $\kappa = 0$ . The best description (curve 2) is obtained for  $\kappa = 1.1$ .

tion that  $\kappa = 1.1$  when  $2\Gamma_k = 23$  kHz. Since the error in the determination of  $\Gamma_k$  was  $\pm 25\%$ , an analysis of the experimental results used in the minimum and maximum values of  $\Gamma_k$  indicated that the error in the determination of  $\kappa$  was  $\pm 35\%$ .

The coefficient  $S_k$  of nonlinear interaction of nuclear spin waves in  $\text{MnCO}_3$ , calculated using Eq. (9) and the experimental value  $\alpha_m \approx 1.2 \times 10^{-5}$  W/Oe at  $T = 2$  K and  $\nu_m = 300$  kHz, gave  $|S_k^{\text{exp}}| \approx 10$  GHz. The high error in the determination of  $S_k$  (up to an order of magnitude) was due to the combination of all the errors in the parameters occurring in Eq. (9). A theoretical calculation of  $S_k$  carried out using Eq. (A3) from Ref. 10 gave  $|S_k^{\text{theor}}| \approx 2.9$  GHz.

Dependences similar to those plotted in Figs. 1–3 were obtained also for  $\text{CsMnF}_3$ . The parameters  $\kappa$  and  $\tau_0^{-1}$  were determined from the experimental data for  $T = 2\text{--}4.2$  K and  $\nu_p = 1000$  MHz and their values were  $\kappa \approx 0.5$  and  $\tau_0^{-1} \approx 1.2$ . An analysis of the experimental results gave  $S_k$  at  $T = 2$  K and  $\nu_p = 1000$  MHz: it amounted to  $|S_k^{\text{exp}}| \approx 1$  GHz, whereas the theoretical value was  $|S_k^{\text{theor}}| \approx 5.1$  GHz. Therefore, the values of  $S_k$  calculated from the experimental data were in order-of-magnitude agreement with the theory.

As reported earlier,<sup>13</sup> in the range of pump frequencies  $\nu_p = 700\text{--}900$  MHz the process of parametric excitation of nuclear spin waves was “hard”: the thresholds for the appearance ( $h_{c1}$ ) and quenching ( $h_{c2}$ ) of a parametric resonance were different ( $h_{c1} > h_{c2}$ ); the ratio  $h_{c1}/h_{c2}$  was highest at  $\nu_p^* \approx 780$  MHz and it decreased rapidly away from this frequency. An investigation of the modulation response showed that the dependences  $\alpha_m(\nu_m)$  obtained for the same values of  $H_0$ ,  $T$ , and  $h/h_{c2}$  had different profiles at the hardness maximum and far from it: the  $\alpha_m(\nu_m)$  curve was strongly broadened at the hardness maximum. The theory made it possible to describe satisfactorily the experimental results on the assumption that the coefficient  $\kappa$  of the positive nonlinear damping increased strongly in the hard excitation region. The error in the determination of the coefficients  $\kappa$  and  $\tau_0^{-1}$  increased on increase in the hardness: for example, the theoretical dependences  $\alpha_m(\nu_m)$  at  $\nu_p = 781$  MHz were practically indistinguishable for  $\kappa = 2$  and  $\kappa = 3$ .

Another interesting feature of the process of parametric excitation of nuclear spin waves in  $\text{CsMnF}_3$  was the anisotropy of the fields  $h_{c1}$  and  $h_{c2}$  when  $H_0$  was rotated in the basal plane of the crystal.<sup>14</sup> As pointed out in Ref. 14, this anisotropy was a manifestation of the corresponding anisotropic dependence<sup>15</sup> of the excitation threshold of electron spin waves in the same crystal. One of the consequences of this anisotropy of the threshold amplitudes (i.e., of the anisotropy of relaxation of electron spin waves) could be a nonisotropic nature of the distribution of parametric magnons in the  $k$  space at low values of the supercriticality.<sup>3)</sup>

The nonisotropic distribution of parametric magnons was supported by a giant anisotropy of the modulation response  $\alpha_m$  when the field  $H_0$  was rotated in the basal plane of a crystal (Fig. 4). It should be pointed out that the behavior of  $\alpha_m(\varphi)$  was correlated with the dependence  $h_{c2}(\varphi)$ , but the degrees of anisotropy  $G_{\alpha_m} \approx 30$  and  $G_{h_{c2}} \approx 0.7$  at  $\nu_p = 780$  MHz differed by more than one order of magnitude:

$$G_A = \max[A(\varphi)] / \min[A(\varphi)] - 1.$$

The anisotropy of the parameters of the steady state were determined by investigating the dependence  $\alpha_m(\nu_m)$  at  $\nu_p = 780$  MHz and  $T = 2$  K when  $(h/h_{c2})^2 = 10$  using two directions of the magnetic field corresponding to the maximum and minimum of the signal  $\alpha_m$ . In the  $\varphi = 30^\circ$  case the  $\alpha_m(\nu_m)$  curve had a wider maximum than for  $\varphi = 0^\circ$ . An analysis of the results yielded the following estimates:

$$\text{for } \varphi = 30^\circ: \tau_0^{-1} \approx 1.5; \kappa \approx 2, |S_k| \approx 50 \text{ GHz},$$

$$\text{for } \varphi = 0^\circ: \tau_0^{-1} \approx 1, \kappa \approx 1, |S_k| \approx 500 \text{ GHz}$$

( $\alpha_m \approx 3 \times 10^{-5}$  W/Oe for  $\nu_m = 100$  kHz and  $\Gamma_k = 15$  kHz). The theoretical value was  $|S_k| = 15$  GHz.

An investigation of the dependence of the anisotropy of the modulation response  $G_{\alpha_m}(\nu_p)$  and of the susceptibility  $G_{\chi''}(\nu_p)$  on the pump frequency showed that  $G_{\alpha_m} \approx G_{\chi''}$  were maximal at the hardness peak and fell rapidly away from  $\nu_p^* = 780$  MHz. For example, at  $\nu_p = 775$  MHz their values decreased approximately threefold, whereas at  $\nu_p = 975$  MHz the reduction was a factor of 50. Such frequency dependences of  $G_{\alpha_m}$  and  $G_{\chi''}$  were correlated with

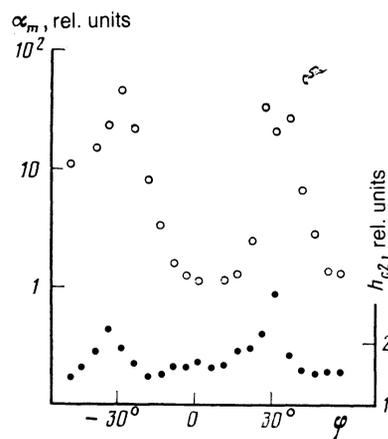


FIG. 4. Anisotropy of the threshold field  $h_{c2}$  and of the modulation response  $\alpha_m(0)$  for the  $nn$  process in  $\text{CsMnF}_3$  on rotation of  $H_0$  in the basal plane of the crystal;  $\varphi = 0$  corresponds to the binary axis of the crystal,  $\nu_p = 779$  MHz,  $\nu_m = 200$  kHz,  $H_0 = 1$  kOe,  $T = 1.85$  K;  $\alpha_m(\varphi)$  was recorded for  $h/h_{c2} = 10$  dB.

the corresponding dependence of the parameter representing the hardness of magnon excitation, whose value  $h_{c1}/h_{c2} - 1$  was maximal at  $\nu_p^* \approx 780$  MHz and fell rapidly when the pump frequency varied in such a way that at  $|\nu_p - \nu_p^*| \gtrsim 100$  MHz this effect was no longer observed.

This relationship was supported also by the observation that the values of the hardness and anisotropy parameters varied in the same way from sample to sample. For example, in the case of  $\text{CsMnF}_3$ , which was the crystal used in our main measurements, it was found that  $G_{\alpha_m} \approx 30$  and  $h_{c1}h_{c2} - 1 \approx 6$  at a frequency  $\nu_p^* \approx 780$  MHz, whereas in the case of the other sample under the same conditions we obtained  $G_{\alpha_m} \approx 7$  and  $h_{c1}/h_{c2} - 1 \approx 2$ . It was demonstrated in Ref. 14 that the hard nature of the excitation of nuclear spin waves in  $\text{CsMnF}_3$  was due to their interaction with dislocations. The observed correlation between the hardness parameter and the anisotropy of  $\alpha_m$  and  $\chi''$  indicated that these phenomena were of common origin.

### ANOMALIES OF MODULATION RESPONSE OF NUCLEAR SPIN WAVES IN $\text{CsMnF}_3$

An investigation of the modulation response of a system of parametric nuclear spin waves in  $\text{CsMnF}_3$  revealed several striking features of the dependences of  $\alpha_m$  on  $\nu_m$  and  $h/h_{c2}$ . One of them, due to excitation of magnetoelastic oscillations of a sample, had been reported earlier.<sup>17</sup> Here, we shall consider a different anomaly observed for supercriticalities  $h/h_{c2} \lesssim 3$  in the range of pump frequencies  $\nu_p = 750$ – $830$  MHz (where parametric excitation of nuclear spin waves was hard) at temperatures  $T \lesssim 2.5$  K. We found (Fig. 5) that an increase in the supercriticality resulted in a major modification of the response of the system of parametric nuclear spin waves. The maximum of the dependence  $\alpha_m(\nu_m)$  shifted toward lower frequencies and in a narrow frequency interval near  $\nu_m \approx 50$  kHz the value of  $\alpha_m$  began to fall rapidly, but the "dip" gradually flattened on further increase in  $h/h_{c2}$ . It should be noted that the dependence  $\alpha_m(h/h_{c2})$  then exhibited hysteresis when the microwave power was increased and then reduced (Fig. 6). The supercriticality  $h/h_{c2}$  at which this anomaly was observed increased on reduction in  $H_0$  and on increase in  $T$ , and when the pump

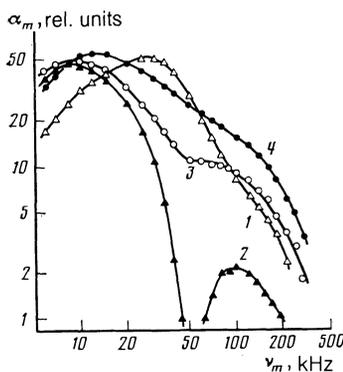


FIG. 5. Frequency dependences of the modulation response of  $\text{CsMnF}_3$  in the case of the  $nn$  process and  $\nu_p = 822$  MHz,  $H_0 = 1$  kOe, and  $T = 1.94$  K obtained for different values of the supercriticality  $h/h_{c2}$ : 1) 2.51; 2) 2.82; 3) 3.16; 4) 3.55. The continuous curves are drawn for clarity.

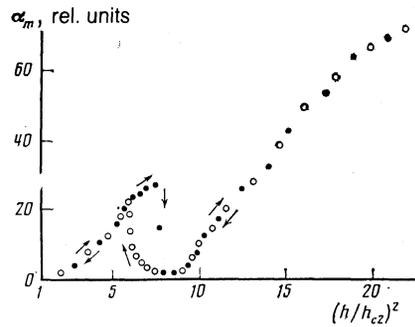


FIG. 6. Dependence of the modulation response on the supercriticality at  $\nu_m = 50$  kHz (the experimental conditions were the same as in Fig. 5). The black circles describe the behavior of  $\alpha_m$  when the supercriticality was increased and the open circles are the results obtained when the supercriticality was reduced.

frequency  $\nu_p$  approached  $\nu_p^* \approx 780$  MHz (corresponding to the maximum hardness of the excitation of nuclear spin waves), this quantity fell to  $h/h_{c2} \approx 1$ . It should be pointed out that the characteristic frequency of the dip in the  $\alpha_m(\nu_m)$  dependence remained practically unaffected.

The existence of this singularity and its behavior were independent of the modulation amplitude  $H_m$ ; this demonstrated that in our case the modulation of the field was simply a probe which enabled us to detect modification of the steady state of parametric nuclear spin waves. Such modification was not manifested in any way in the behavior of the dynamic nonlinear susceptibility  $\chi''$ .

At present we are unable to provide an explanation of the observed effect, but we can put forward some suggestions which might help in the interpretation. Our  $\text{CsMnF}_3$  sample was a plane-parallel plate of thickness  $d = 0.9$  mm. An estimate of the reciprocal of the travel time of a nuclear magnon between the boundaries of a sample obtained from  $t_n^{-1} = v_n/d$  ( $v_n$  is the group velocity of nuclear spin waves) was in good agreement with the characteristic frequency of the  $\approx 50$  kHz anomaly. This circumstance suggested a characteristic size effect in the nuclear subsystem. An increase in  $T$  and a reduction in  $H_0$  increased the rate of relaxation of nuclear spin waves and this reduced their mean free path, so that the size effect should disappear.

### STEADY STATE OF A SYSTEM OF PARAMETRIC ELECTRON SPIN WAVES

We shall now consider the results of measurements of the modulation response  $\alpha_m$  of parametric electron spin waves in  $\text{MnCO}_3$  and  $\text{CsMnF}_3$ . It should be pointed out immediately that the process of parametric excitation of electron spin waves in these antiferromagnets was hard.<sup>18,19</sup> Therefore, one could hardly count on a detailed theoretical description of the results obtained. The dependences  $\alpha_m(\nu_m, h/h_{c2})$  were similar for both crystals: the  $\alpha_m(\nu_m)$  curves were in the form of a wide plateau with a characteristic fall at low and high modulation frequencies (Fig. 7). It was interesting to note that a similar dependence  $\alpha_m(\nu_m)$  had been obtained also for nuclear spin waves in the region of their hard excitation.

Figure 7 includes also a theoretical curve plotted on the basis of Eq. (9) on the assumption that  $\kappa = 0.2$ ,  $\tau_0^{-1} = 0.1$ , and  $\Gamma_k = 0.1$  MHz. The selected value of the parameter  $\kappa$

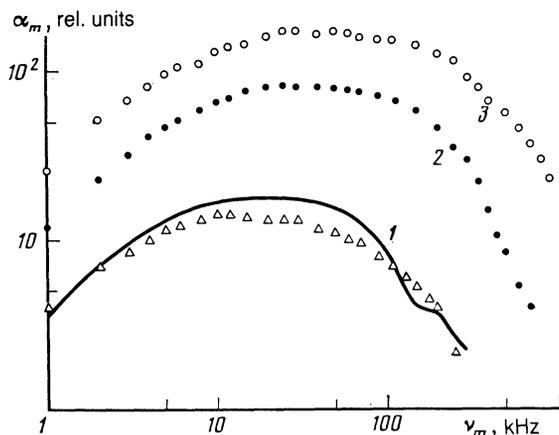


FIG. 7. Frequency dependences of the modulation response of  $\text{CsMnF}_3$  in the case of the  $ee$  process for  $\nu_p = 22.7$  GHz,  $H_0 = 3$  kOe,  $T = 1.65$  K, and the following values of the supercriticality  $h/h_{c2}$ : 1) 1.023; 2) 1.12; 3) 2.88. The continuous curve is calculated for  $\tau_0^{-1} = 0.1$ ,  $\kappa = 0.2$ , and  $\Gamma_k = 100$  Hz.

was deduced from an analysis of the data for the nonlinear dynamic susceptibility  $\chi'$  and  $\chi''$  (Ref. 4) (which gave  $\kappa = 0.2 \pm 0.1$ ), and  $\Gamma_k = 0.1 \pm 0.05$  MHz was deduced from the parametric excitation threshold.

## CONCLUSIONS

1. Determination of the modulation response provides a simple and quite informative method for investigating the characteristics of a steady state of a system of parametric spin waves. This method makes it possible to observe features of the behavior of the system above the parametric excitation threshold which are not revealed by a study of the nonlinear dynamic susceptibility of a magnetic material.

2. A theoretical expression for the modulation response obtained allowing for two mechanisms limiting the amplitude of parametric spin waves, which are the phase mechanism and the positive nonlinear damping, provides a satisfactory description of the experimental results obtained in a wide range of supercriticalities (excess above the threshold).

3. A giant hexagonal anisotropy of the modulation response and of the nonlinear dynamic susceptibility  $\chi''$  exhibited by  $\text{CsMnF}_3$  on excitation of nuclear spin waves may be

an indication of an anisotropic distribution of parametric magnons in the  $k$  space.

4. A correlation of the anisotropy of  $\alpha_m$  and  $\chi''$  with the hardness parameter  $h_{c1}/h_{c2} - 1$  is evidence of the common origin of these effects which are due to the interaction of nuclear magnons with defects in a crystal.

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<sup>11</sup>It should be pointed out that in the case of these contributions the average over a period  $2\pi/\omega_m$  is zero, i.e., they do not allow for the influence of the power absorbed from the modulation field.

<sup>12</sup>We shall assume that the criterion of stability of the steady state is satisfied.<sup>12</sup>

<sup>13</sup>The possibility of nonisotropic distributions of parametrically excited spin waves in antiferromagnetics was pointed out in the theoretical treatments in Refs. 3 and 16.

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