

# Investigation of a combined resonance of mixed parametric magnon pairs in antiferromagnetic $\text{MnCO}_3$

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The threshold fields for the excitation of electron ( $e$ ) and nuclear ( $n$ ) spin waves, of frequencies satisfying the combined resonance condition, were determined for antiferromagnetic  $\text{MnCO}_3$ . This made it possible to check the expression describing the parametric threshold of the combined  $en$  process. The influence of modulation of a static magnetic field on the combined resonance threshold was studied and the experimental results were explained satisfactorily. The dependences of the modulation response of a system of  $en$  pairs on the excess above the critical excitation and on the modulation frequency were determined. A theory of a stationary state of magnons excited by  $en$  pumping was developed and used to calculate the susceptibility and the modulation response. The ranges of the parameters in which this theory explained satisfactorily the experimental results were identified.

## INTRODUCTION

A parametric resonance of spin waves in microwave fields provides a very convenient method for investigating the transport properties of magnetic materials and modeling the effects occurring in a nonlinear magnetic medium and associated with the interaction between finite-amplitude waves and such a medium. Among the most interesting objects for such investigations are weakly anisotropic antiferromagnets ( $\text{RbMnF}_3$ ) and antiferromagnets with the easy-plane magnetic anisotropy ( $\text{MnCO}_3$ ,  $\text{CsMnF}_3$ ,  $\text{CsMnCl}_3$ ). They have a branch of "electron" ( $e$ ) spin waves (of relatively low frequencies  $\nu_{ek} \sim 10\text{--}100$  GHz), a branch of "nuclear" ( $n$ ) spin waves ( $\nu_{nk} \sim 0.2\text{--}0.7$  GHz), and acoustic phonons ( $ph$ ). The interaction with an external field  $h \cos(2\pi\nu_p t)$  may give rise to various types of parametric instability  $\nu_p = \nu_{\alpha k} + \nu_{\beta - k}$  when a microwave field photon splits into two quasiparticles  $\alpha$  and  $\beta$  ( $\alpha, \beta = e, n, ph$ ) with equal and oppositely directed wave vectors (see Ref. 1). The most thoroughly investigated parametric instabilities are those of a "degenerate" resonance type characterized by  $\alpha\beta \equiv ee$  and  $\alpha\beta \equiv nn$ ; the threshold fields  $b_c^{ee}$  (Refs. 2 and 3) and  $h_c^{nn}$  (Refs. 4 and 5) and states above the thresholds ( $ee$  process<sup>6,7</sup> or  $nn$  process<sup>8,10</sup>) of such stabilities have been investigated quite thoroughly. Much less work has been done on combined parametric instabilities such as those of the  $\alpha\beta \equiv en$  type when an excited pair consists of one electron magnon and one nuclear magnon.<sup>1,2,11,12</sup>

Our aim was to investigate the threshold and the stationary state beyond the threshold of the  $en$  process. The results are presented in the following order. The first section describes the experimental method. The second section reports a study of the threshold of the  $en$  process and the third deals with the influence of an external magnetic rf field on this threshold. The fourth and fifth sections give the results of an investigation of a system of parametrically excited  $en$  pairs of magnons in a state above the threshold. The main results of the study are formulated in the section headed Conclusions.

## 1. METHOD

Parallel pumping ( $\mathbf{h} \parallel \mathbf{H}$ ) creating pairs of electron and nuclear magnons was carried out at a frequency  $\nu_p \approx 9.3$

GHz at temperatures  $T = 1.55\text{--}2.17$  K. The source of microwave power was a magnetron which provided continuously an output power of  $\sim 10$  W. Our measurements were usually carried out under pulse modulation conditions when the microwave power was modulated by varying the voltage supplied to the magnetron or using a pin diode. The duration of the pulses was  $100\text{--}1000 \mu\text{s}$  and the repetition frequency was 50 Hz.

A calibrated attenuator and a circulator were used to deliver microwave pulses to a rectangular resonator with a  $Q$  factor  $Q \approx 1500$  under load. A sample of  $\text{MnCO}_3$  of  $4 \times 3 \times 0.5 \text{ mm}^3$  dimensions was bonded to the bottom of this resonator. A signal reflected (or transmitted) by the resonator was detected and observed on the screen of an oscilloscope. The coupling of the resonator to the input waveguide was near-critical. The onset of the  $en$  process was deduced from the appearance of a sharp dip in the microwave pulse transmitted by the resonator.

Modulation of the magnetic field ( $\mathbf{H}_m \parallel \mathbf{H}$ ) of  $H_m \leq 0.5$  Oe amplitude and  $\nu_m = 1 \text{ kHz--}4 \text{ MHz}$  frequency was imposed by a loop which could also be used to raise the temperature of the nuclear subsystem in a sample (by heating with the rf field) relative to the temperature of the helium bath. In the latter case the  $\mathbf{h} \parallel \mathbf{H} \perp \mathbf{H}_{rf}$  geometry was used, which ensured an increase in the temperature of nuclear spins by the method described in Ref. 13.

The amplitude of fluctuations of the microwave power absorbed by a sample was measured with a selective rf microvoltmeter of the V6-1 type, which determined the modulation  $\Delta U$  of the voltage across a crystal detector receiving the microwave signal transmitted by the resonator. The power  $P_{\text{det}}$  incident on the detector was assumed to be proportional to the difference between the powers reaching the resonator and that absorbed by the sample. The appearance of amplitude modulation  $\Delta P \cos(2\pi\nu_m t)$  in the absorbed microwave power gave rise to amplitude modulation of the detector signal and its depth was proportional to  $\Delta P$ . A decoupling attenuator was used to ensure that the microwave field amplitude reaching the detector was such that the diode operated as a square-law detector of  $h$ , i.e.,  $U_{\text{det}} = DP_{\text{micr}}$ . The microwave signal was detected using a D608 mixing diode which produced a signal characterized by  $\Delta U = D\Delta P \cos(2\pi\nu_m t)$  throughout the range of modulation frequencies used in our

experiments ( $\nu_m \leq 4$  MHz). The amplitude of the modulation field  $H_m$  was selected so that the influence of  $H_m$  on the pump threshold was less than 1% and the value of  $\Delta P$  was proportional to  $H_m$ , i.e., a linear modulation response was obtained. The methods used in the determination of the thresholds of the  $ee$  and  $nn$  processes was described in Refs. 3 and 5, respectively.

## 2. THRESHOLD OF THE $en$ PROCESS

Combined excitation of electron and nuclear magnons was one of the first parametric processes detected experimentally in antiferromagnets.<sup>11</sup> However, the studies of the process did not move much beyond the determination of the threshold amplitude  $h_c^{en}$ . This was due to the fact that the  $en$  process was not very informative in studies of relaxation of excited magnons, because the expression for the threshold contains a product of the relaxation rates of electron and nuclear magnons<sup>1,11</sup>:

$$h_c^{en} = (h_c^{ee} h_c^{nn})^{1/2} \propto (\Gamma_{ek} \Gamma_{nk})^{1/2}. \quad (1)$$

According to the conclusions reached in Refs. 1, 2, and 12, calculations of  $\Gamma_{nk}$  carried out using the threshold  $h_c^{en}$  and  $h_c^{ee}$  gave values of the rate of relaxation of nuclear spin waves which did not agree with the results of direct calculation of  $\Gamma_{nk}$  on the basis of the degenerate  $nn$  process. This raised doubts about the validity of the theoretical expression for  $h_c^{en}$  given above.

It is now clear that the large discrepancies between the calculated and experimental values of  $h_c^{en}$  reported in Refs. 1, 2, and 12 were due to errors in the determination of the absolute value of the threshold  $h_c^{ee}$ , and an incorrect estimate of  $h_c^{nn}$ , which depended strongly on the frequency of excited nuclear magnons. For example, the value of  $h_c^{en}$  was estimated in Ref. 1 using the threshold  $h_c^{nn}$  measured for nuclear magnons of frequency  $\nu_{nk} = 437$  MHz, whereas the  $en$  process excited nuclear spin waves with  $\nu_{nk} = 520$  MHz for which the threshold was an order of magnitude higher. Therefore, a reliable check of Eq. (1) required measurements of the threshold field of the  $en$ ,  $ee$ , and  $nn$  processes under conditions such that degenerate pumping excited the same quasiparticles as nondegenerate pumping.

We carried out such measurements on antiferromagnetic  $\text{MnCO}_3$ . The threshold field of the  $en$  process in our sample observed when the pump frequency was  $\nu_p = 9.35$  GHz amounted to  $h_c^{en} = 0.18 + 0.04$  Oe at  $T = 1.7$  K when the field was  $H = 0.75$  kOe; this resulted in excitation of electron and nuclear magnons of frequencies 8.83 GHz and 520 MHz, respectively. The product of the threshold fields for the excitation of the  $ee$  and  $nn$  processes was found to be  $(h_c^{ee} h_c^{nn})^{1/2} = 0.16 \pm 0.04$  Oe. Therefore, for these values of the parameters equation (1) was satisfied within the limits of the experimental error.

The next stage in checking the validity of Eq. (1) involved a study of its functional dependence. The complete expression for the threshold  $h_c^{en}$  is<sup>1</sup>

$$h_c^{en} = 4 \left( \frac{\pi \hbar}{\mu_B} \right)^3 \frac{(\Gamma_{ek} \Gamma_{nk})^{1/2}}{H_\Delta} \frac{\nu_{ek}^{3/2} \nu_{nk}^{1/2}}{\nu_n (2H + H_D)}. \quad (2)$$

Here,  $H_\Delta^2 = 5.8/T$  [kOe<sup>2</sup>] is the hyperfine interaction parameter;  $\nu_n = 640$  MHz is the Larmor NMR frequency;  $H_D$

$= 4.4$  kOe is the Dzyaloshinskii field. Our experiments were carried out at a fixed pump frequency  $\nu_p = 9.35$  GHz. It was difficult to investigate the dependences of  $h_c^{en}$  on the static magnetic field  $H$  under these conditions because of the presence of the magnon-phonon peak in a field of  $H \approx 1$  kOe (Ref. 1), so that the most informative dependences were those of  $h_c^{en}$  on  $T$  and  $T_n$ , where  $T_n$  is the temperature of the subsystem of nuclear spins. Qualitatively different results might be expected when these parameters are altered. In the former case the temperatures of the electron and nuclear subsystems varied in a similar manner ( $T = T_n$ ) whereas in the latter case we found that  $T \neq T_n$ , i.e., the equilibrium populations of electron and nuclear spin waves obeyed different laws. In the case of the  $en$  process we obtained  $\nu_{ek} \approx \text{const}$ , whereas  $\nu_{nk}$  was not greatly affected by the variation of  $T_n$ . Therefore, allowing for the dependence  $H_\Delta \propto N_n^{-1/2}$ , governing the threshold  $h_c^{en}$ , we obtained

$$h_c^{en} \propto T_n^{1/2} [\Gamma_{ek}(T, T_n) \Gamma_{nk}(T, T_n)]^{1/2}. \quad (3)$$

Figure 1 shows the results of measurements  $h_c^{en}$  at temperatures  $T = 1.55$ – $2.15$  K and  $T_n = 1.55$ – $2.35$  K. At these temperatures in fields  $H \leq 1$  kOe the rate of relaxation of electron magnons was known to be independent of  $T$  (Ref. 3) or weakly dependent on  $T$  (Ref. 12), and the rate of relaxation of nuclear magnons was directly proportional to  $T_n$  (Refs. 4 and 15). Consequently, we could reduce Eq. (3) to  $h_c^{en} \propto T_n$ . This dependence for the threshold of the  $en$  process described quite satisfactorily the experimental results presented in Fig. 1. Therefore, the theoretical value  $h_c^{en}$  given by Eq. (2) was in agreement with the experimental data not only in magnitude, but also of the functional dependence.

We shall complete this section by discussing the causes of "soft" excitation of magnons in a combined process. Parametric excitation of electron magnons in the  $ee$  process is known to be "hard" (Refs. 16 and 17), i.e., it is characterized by the appearance threshold  $h_{c1}$  and by the quenching threshold  $h_{c2}$  of the parametric instability ( $h_{c1} > h_{c2}$ ). This effect can be explained phenomenologically by negative nonlinear magnon damping. In the case of nondegenerate pumping such negative nonlinear damping of electron magnons is clearly compensated by positive nonlinear damping due to interaction with parametric nuclear spin waves. Under our experimental conditions the main contribution to relaxation

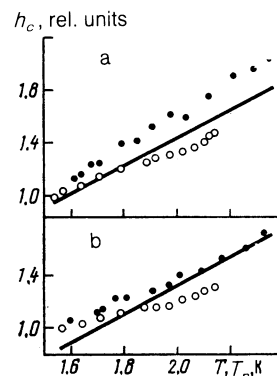


FIG. 1. Dependence of the threshold of the  $en$  process on the temperatures  $T$  (O) and  $T_n$  (●) in fields  $H = 0.5$  kOe (a) and  $0.75$  kOe (b).

of parametric electron and nuclear magnons comes from the processes of elastic scattering by fluctuations of the longitudinal magnetization of the subsystem.<sup>3,4</sup> For this reason the number of electron and nuclear magnons with wave vectors equal in absolute value to the wave vectors of excited magnons increases strongly above the threshold. The result is strong enhancement of the scattering processes, such that

$$e(\mathbf{k}) + n(\mathbf{q}) \rightarrow e(\mathbf{q}) + n(\mathbf{k}),$$

when electron and nuclear magnons exchange momenta. Positive nonlinear damping due to these (and analogous) processes can balance negative nonlinear damping of electron spin waves. Therefore, the excitation process becomes soft. It would be interesting to check this hypothesis by experiments involving simultaneous excitation of the  $ee$  and  $nn$  processes in which the excited electron and nuclear spin waves have the same values of  $|\mathbf{k}|$ . In an experiment of this kind the pumping of nuclear spin waves should significantly affect the threshold field  $h_c^{ee}$ .

### 3. INFLUENCE OF A RADIOFREQUENCY FIELD ON THE THRESHOLD OF THE $en$ PROCESS

We now consider the influence of an external oscillating magnetic field  $H_m \cos(2\pi\nu_m t)$ , in the case when  $\mathbf{H}_m \parallel \mathbf{h}$  and  $\nu_m \ll \nu_p$ , on the threshold of the parametric  $en$  process. The possibility of an increase in the threshold amplitude of a "degenerate" parametric resonance of magnons by modulation of the external magnetic field was first pointed out by Suhl.<sup>18</sup> An increase in  $h_c$  occurs because of modulation of the magnon spectrum, as a result of which the parametric resonance condition varies periodically. The theory of Suhl<sup>18</sup> appeared at about the same time as an experimental paper<sup>19</sup> reporting the first observation of an increase in the threshold of the  $ee$  pumping of yttrium iron garnet (YIG) under field modulation conditions. Subsequent detailed investigations were reported in Ref. 20 ( $ee$  process in YIG), in Ref. 21 ( $ee$  process in antiferromagnets  $\text{MnCO}_3$  and  $\text{CsMnF}_3$ ), and in Refs. 22–24 ( $nn$  process in  $\text{CsMnF}_3$ ). A satisfactory agreement between the theory and experiment was achieved in all these investigations.

We consider the influence of a modulating field on the nondegenerate parametric process threshold. A calculation based on the equations of motion for the intensities of the excited waves (these equations will be given explicitly in the next section) leads to the expression ( $\nu_m \gtrsim \Gamma_{ek} + \Gamma_{nk}$ )

$$\frac{h_c}{h_{c0}} - 1 = \frac{1}{4} \left( \frac{\partial \nu_{ek}}{\partial H} + \frac{\partial \nu_{nk}}{\partial H} \right)^2 \frac{H_m^2}{(\Gamma_{ek} + \Gamma_{nk})^2 + \nu_m^2}. \quad (4)$$

Here  $h_c/h_{c0}$  denoted the relative increase in the threshold,

$$\frac{\partial \nu_{ek}}{\partial H} = \left( \frac{\mu_B}{\pi \hbar} \right)^2 \frac{2H + H_D}{2\nu_{ek}}, \quad (5)$$

$$\frac{\partial \nu_{nk}}{\partial H} = \frac{\nu_n^2}{\nu_{nk} \nu_{ek}} \left( \frac{\mu_B}{\pi \hbar} \right)^2 \left( \frac{H_\Delta}{\nu_{ek}} \right)^2 \frac{\partial \nu_{ek}}{\partial H}.$$

It should be pointed out that the replacement  $n \rightarrow e$  or its opposite reduces Eq. (4) to the expression for the relative increase in the threshold as a result of  $ee$  (Ref. 21) or  $nn$  (Ref. 24) pumping. It is interesting to note that in the limit of high frequencies  $\nu_m \gg \Gamma_{ek}$  the influence of modulation on the nondegenerate pumping threshold differs, in accordance

with Eq. (4) and the inequality  $\partial \nu_{ek} / \partial H \gg \partial \nu_{nk} / \partial H$ , only by the factor 1/4 from the corresponding expression for the  $ee$  pumping case.

Figures 2 and 3 show the experimental results of an investigation of the influence of the modulation field on the  $en$  process threshold. As expected, for the same values of the parameters the influence of modulation on the threshold amplitude of the  $en$  process was less than in the degenerate pumping case. It is clear from Fig. 3 that the theoretical curve described satisfactorily, without any fitting parameters, the experimental dependence obtained in the frequency range  $\nu_m \gtrsim 2\Gamma_{ek} \approx 0.8$  MHz. The functional dependence of the threshold on the amplitude of the modulation field obeyed the law  $(h_c/h_{c0} - 1) \propto H_m^2$  up to a frequency  $\nu_m = 500$  kHz (Fig. 2). At a lower frequency of  $\nu_m = 300$  kHz the  $H_m^2$  law was disobeyed even in fields  $H_m \gtrsim 0.2$  Oe. This agreement between the theory and experiments could be regarded as satisfactory, because Eq. (4) was obtained in the  $\nu_m \gtrsim \Lambda_{ek} + \Gamma_{nk}$  approximation.

### 4. STATIONARY STATE ABOVE THE THRESHOLD

We shall analyze the state of a system of  $en$  pairs above the threshold using a model of a single-frequency turbulence considered by L'vov and Rubenchik in Ref. 25 in terms of the classical Hamiltonian formalism for waves in a plasma. The interaction Hamiltonian in this model is diagonal in the wave pairs and the equations of motion are written down for correlation functions

$$N_{ek} \equiv \langle a_{ek}^* a_{ek} \rangle, \quad N_{nk} \equiv \langle a_{nk}^* a_{nk} \rangle, \quad \sigma_k \equiv \langle a_{ek} a_{n-k} \exp(-i\omega_p t) \rangle,$$

where  $a_{jk}^*$  and  $a_{jk}$  are the complex amplitudes of waves and the averaging is carried out over the individual phases of the waves (here and later we have  $\omega \equiv 2\pi\nu$  and  $\gamma \equiv 2\pi\Gamma$ ). It is also assumed that only those pairs are excited which are most strongly coupled to the pumping, and in the case of a stationary state these are the waves satisfying the "resonance" surface condition :

$$\Delta_k = \omega_p - \tilde{\omega}_{ek} - \tilde{\omega}_{nk} = 0, \quad (6)$$

where

$$\tilde{\omega}_{ek} = \omega_{ek} + 2\mathcal{F}_e(k) N_{ek} + \mathcal{F}_{en}(k) N_{nk},$$

$$\tilde{\omega}_{nk} = \omega_{nk} + 2\mathcal{F}_n(k) N_{nk} + \mathcal{F}_{en}(k) N_{ek}$$

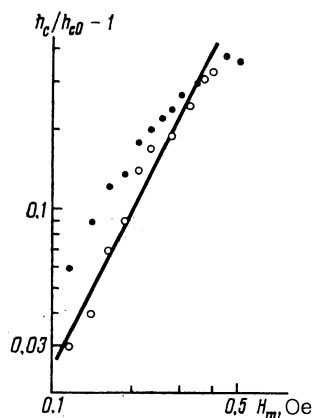


FIG. 2. Dependence of the relative changes in the threshold field of the  $en$  process on the amplitude  $H_m$  when  $\nu_m = 0.3$  MHz (●),  $\nu_m = 0.5$  MHz (○),  $H = 0.6$  kOe and  $T = 1.74$  K. The straight line represents proportionality to  $H_m^2$ .

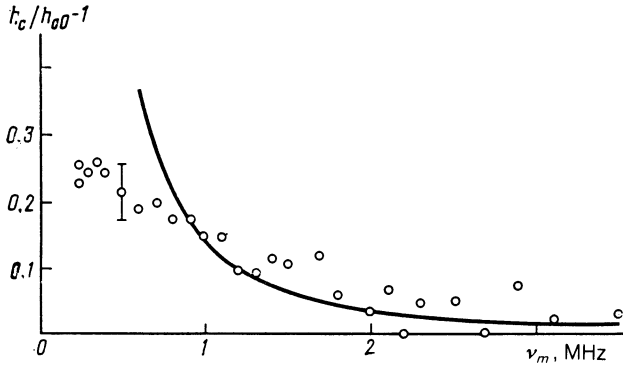


FIG. 3. Dependence of the relative increase of the threshold amplitude of the  $en$  process on the field modulation frequency in the case when  $H_m = 0.3$  Oe,  $H = 0.6$  kOe,  $T = 1.8$  K, and  $\nu_p = 9.32$  GHz. The continuous curve represents the theoretical dependence given by Eq. (4).

are respectively the nonlinearly renormalized frequencies of electron and nuclear spin waves, and the  $\mathcal{F}(k)$  are the interaction amplitudes.

Since in the case of weakly anisotropic antiferromagnets the frequencies and coefficients describing the nonlinear interactions of spin waves are practically independent of the wave vector directions,<sup>26</sup> the resonant surface is a sphere in the  $\mathbf{k}$  space. For simplicity, we shall drop the index  $k$  in most cases.

According to the theory of Ref. 25, the greatest interest lies in the solutions characterized by a strong phase correlation between the waves in a pair:  $|\sigma|^2 = N_e N_n$ . Introducing

$$\sigma = (N_e N_n)^{1/2} \exp[i(\pi/2 - \theta)],$$

where  $\theta$  is the phase of the "mismatch" between the pump field and forced oscillations of the magnetic medium, we find that the equations of Ref. 25 yield the following:

$$\left(\frac{1}{2} \frac{d}{dt} + \gamma_e\right) N_e = \left(\frac{1}{2} \frac{d}{dt} + \gamma_n\right) N_n = (N_e N_n)^{1/2} hV \cos \theta, \quad (7)$$

$$(N_e N_n)^{1/2} \left(\frac{d}{dt} \theta - \Delta\right) + (N_e + N_n) [hV \sin \theta + \mathcal{F}(N_e N_n)^{1/2}] = 0, \quad (8)$$

where  $V$  is a coefficient representing the coupling of parametric pairs with the pump field;  $\mathcal{F}$  is the nonlinear magnon-magnon interaction coefficient.

Equations of the form (7) and (8) can be obtained (under the above assumptions) by a quantum-mechanical approach using the control equation method developed by Lax.<sup>27</sup> We have to assume that the dynamics of a parametric system is governed entirely by pairs of electron and nuclear spin waves which are in exact resonance with the pumping ("dynamic condensate") and the other degrees of freedom of a crystal (other magnons, phonons) form a thermostat. Then, the relaxation rates  $\gamma_e$  and  $\gamma_n$  can be calculated using the known correlations of the thermostat.

Assuming that the time derivatives in Eqs. (7) and (8) vanish, we obtain a system of equations from which we can find the parameters of a stationary state:

$$\gamma_e N_e = \gamma_n N_n = (N_e N_n)^{1/2} hV \cos \theta, \quad (9)$$

$$hV \sin \theta + \mathcal{F}(N_e N_n)^{1/2} = 0. \quad (10)$$

The relationship (9) is equivalent to a rule, known from the theory of combined parametric processes, governing the distribution of the dissipated pump power  $W_p$  among the excited modes<sup>28</sup>:

$$W_p = W_e + W_n, \quad W_e/\omega_e = W_n/\omega_n,$$

where

$$W_m = 2\hbar\gamma_m\omega_m N_m, \quad m=e, n. \quad (11)$$

We consider the stability of a stationary state in the presence of small deviations  $\delta N_e \delta N_n \delta \theta \propto \exp(\lambda t)$ . An analysis of the linearized equations (7) and (8) gives the following cubic equation for the characteristic numbers  $\lambda$ :

$$\sum_{j=0}^3 \mathcal{A}_j \lambda^j = 0, \quad (12)$$

where

$$\begin{aligned} \mathcal{A}_3 &= 1, & \mathcal{A}_2 &= g_e + g_n + g_{en}, \\ \mathcal{A}_1 &= (g_e + g_n)g_{en} + 2f + r_e + r_n, \\ \mathcal{A}_0 &= 2(fg_{en} + r_e G_n + r_n G_e). \end{aligned}$$

Here we have adopted the notation

$$\begin{aligned} g_m &= \gamma_m + 2N_m \partial \gamma_m / \partial N_m \quad (m=e, n), & g_{en} &= \gamma_e + \gamma_n, \\ f &= (\gamma_e \partial \gamma_n / \partial N_n - \gamma_n \partial \gamma_e / \partial N_e) N_n + (\gamma_n \partial \gamma_e / \partial N_e - \gamma_e \partial \gamma_n / \partial N_n) N_e, \\ r_e &= 2\mathcal{F} N_e N_n [2T_e + T_{en} + \mathcal{P}(1 + N_e/N_n)], \\ G_e &= \gamma_e + N_e \partial \gamma_e / \partial N_e - N_n \partial \gamma_n / \partial N_e, \\ r_n &= r_{\left(\begin{smallmatrix} e \rightarrow n \\ n \rightarrow e \end{smallmatrix}\right)}, & G_n &= G_{\left(\begin{smallmatrix} e \rightarrow n \\ n \rightarrow e \end{smallmatrix}\right)}. \end{aligned}$$

The stability of a stationary state requires that the real part of the roots of the characteristic equation (12) be negative. It then follows from the Hurwitz stability criterion (see Ref. 29) that

$$\mathcal{A}_2 > 0, \quad \mathcal{A}_0 > 0, \quad \mathcal{A}_1 \mathcal{A}_2 - \mathcal{A}_0 > 0. \quad (13)$$

In the absence of nonlinear damping, these inequalities become

$$\gamma_e + \gamma_n > 0, \quad (14a)$$

$$\mathcal{P}\mathcal{F} > 0, \quad (14b)$$

$$N_e^2 \mathcal{P}\mathcal{F} + \gamma_n (\gamma_e + \gamma_n)^2 / 2\gamma_e > 0, \quad (14c)$$

where

$$\mathcal{F} = 2\mathcal{F}_e \gamma_n + 2\mathcal{F}_n \gamma_e + (\mathcal{F}_{en} + 2\mathcal{P})(\gamma_e + \gamma_n),$$

$$\mathcal{F}_{..} = 2\mathcal{F}_e \gamma_e + 2\mathcal{F}_n \gamma_n + [\mathcal{F}_{en} + \mathcal{P}(\gamma_e/\gamma_n + \gamma_n/\gamma_e)](\gamma_e + \gamma_n).$$

The inequality (14a) is valid always, whereas the other two inequalities depend on the coefficients  $\mathcal{F}$  and  $\mathcal{F}_{..}$ . We now write down the explicit form of these coefficients<sup>30,31</sup>:

$$\mathcal{F}_e \approx -\omega_E [\omega_{e0}^2 + 3(2\mu_B H/\hbar)^2] / 4\omega_{ek}^2 \mathcal{N}, \quad (15)$$

$$\mathcal{F}_{..} \approx -\omega_E \omega_N^2 / 2\omega_{ek}^2 \mathcal{N}, \quad \mathcal{F}_{en} = \mathcal{P} \approx -\omega_E \omega_N / 2\omega_{ek} \mathcal{N},$$

where  $\omega_E \equiv J_0/\hbar$  is the exchange constant,  $\omega_N$  is the unshift-

ed NMR frequency; and  $\mathcal{N}$  is the number of unit cells in a crystal.

We can therefore readily show that the inequalities (14b) and (14c) are satisfied subject to the conditions in the system (15), i.e., a stationary state for the  $en$  process is stable. It should be added that allowance for positive nonlinear damping in the case of electron and nuclear spin waves does not disturb this stability.

Up to now the state of a system of mixed parametric pairs has been investigated experimentally beyond the threshold only once.<sup>1</sup> The dependences of the susceptibility  $\chi''$  on the excess above the threshold  $h/h_c$  were obtained for  $en$  and  $eph$  processes in a sample of  $\text{MnCO}_3$ . The results taken from that paper are plotted in Fig. 4 using coordinates convenient for a comparison with theory. By definition, the imaginary part of the susceptibility is proportional to the absorbed power. Since in stationary states the absorbed and dissipated microwave powers are equal, it follows that the expression for  $\chi''$  can be represented in the form

$$\chi'' = \frac{2 W_p 1}{\omega_p h^2 \mathcal{V}^2}, \quad (16)$$

where  $\mathcal{V}$  is the volume of the sample. In the case of the phase mechanism of limitation on the rate of excitation of waves, Eq. (16) together with Eq. (11) become

$$\chi'' = \frac{4\hbar V^2}{\mathcal{V}^2 |\mathcal{P}|} \left[ \left( \frac{h}{h_c} \right)^2 - 1 \right]^{1/2} \left( \frac{h}{h_c} \right)^{-2}. \quad (17)$$

We give also the expression for  $\chi''$  in the case when the dominant limitation mechanism is nonlinear damping, i.e., when the rate of relaxation of electron spin waves is  $\gamma_e = \gamma_{e0} + \eta N_n$ , and  $\gamma_n$  is independent of  $N_e$  or  $N_n$ :

$$\chi'' = \frac{4\hbar V^2}{\mathcal{V}^2 \eta} \left[ \left( \frac{h}{h_c} \right)^2 - 1 \right] \left( \frac{h}{h_c} \right)^{-2}. \quad (18)$$

The straight lines labeled 1 are plotted in Fig. 4 using Eq. (17) and assuming that  $\hbar V^2/\mathcal{V}^2 |\mathcal{P}| \approx 4.7 \times 10^{-3}$ . We can

see that these lines are located close to the points for the  $eph$  process. The rise of the points representing the  $en$  process is slower: it is closer to curve 2 plotted using Eq. (18), where  $\hbar V^2/\mathcal{V}^2 |\eta| \approx 3.2 \times 10^{-3}$ . Since in the case of the  $en$  process we have<sup>1</sup>

$$V_{en} = 2 \left( \frac{\mu_B}{\hbar} \right)^3 \frac{(2H+H_D)H_\Delta}{\omega_{eh}^2} \frac{\omega_N}{(\omega_{nh}\omega_{eh})^{1/2}}, \quad (19)$$

we readily obtain an estimate  $\mathcal{V}\eta \approx 8 \times 10^{-13} \text{ cm}^3/\text{s}$ .

A theoretical estimate of the parameter  $\hbar/V^2/\mathcal{V}^2 |\mathcal{P}|$  for the  $eph$  process could be obtained assuming again that in antiferromagnets the amplitudes of nonlinear magnetoelastic oscillations are governed primarily by nonlinearities of the magnetic system.<sup>32</sup> In fact this means that

$$V_{eph} \propto V_{ee} \mathcal{B}, \quad \mathcal{P}_{eph} \propto \mathcal{P}_{ee} \mathcal{B}^2,$$

where the parameter  $\mathcal{B}$  represents the linear magnetoelastic coupling. We therefore find that

$$V_{eph}/|\mathcal{P}_{eph}| \sim V_{ee}/|\mathcal{P}_{ee}|.$$

Since it follows from Ref. 30 that

$$V_{ee} \approx \left( \frac{\mu_B}{\hbar} \right)^2 \frac{2H+H_D}{\omega_{eh}}, \quad \mathcal{P}_{ee} = \mathcal{F}_e, \quad (20)$$

we can see that for characteristic values of the experimental parameters<sup>1</sup> which are  $\mathcal{V} \sim 10^{-2} \text{ cm}^3$  and  $\mathcal{N} = \mathcal{V}/\mathcal{V}_0 \sim 10^{20}$  ( $\mathcal{V}_0$  is the volume of a unit magnetic cell), we obtain

$$\hbar V_{eph}/\mathcal{V}^2 |\mathcal{P}_{eph}| \sim 10^{-5} - 10^{-3},$$

which together with the qualitative nature of the discussion can be regarded as in satisfactory agreement with the above value.

## 5. MODULATION RESPONSE

One of the most convenient methods for the investigation of a stationary state of parametrically excited spin

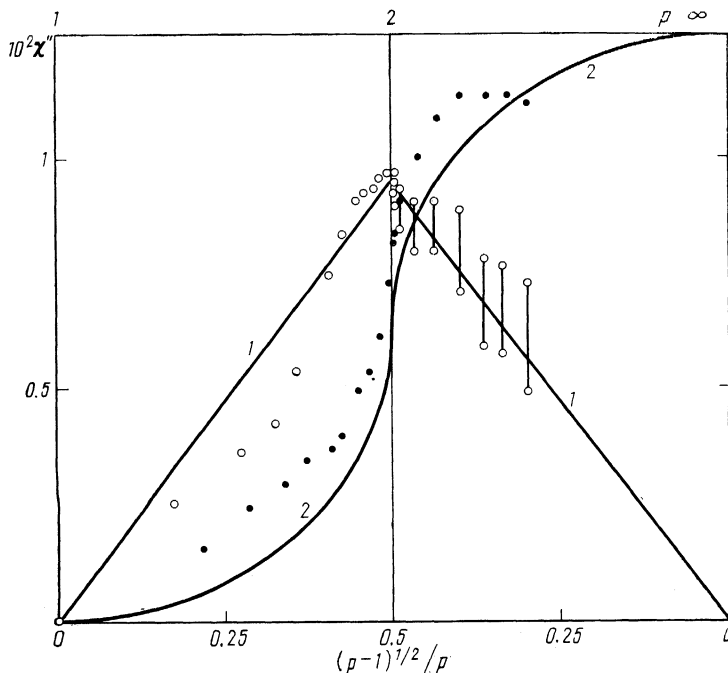


FIG. 4. Behavior of the susceptibility  $\chi''$  above the threshold in the case of the  $en$  ( $\bullet$ ) and  $eph$  ( $\circ$ ) processes;  $\nu_p \approx 9.3 \text{ GHz}$ ,  $T = 1.7 \text{ K}$ , and  $H = 0.5 \text{ kOe}$  (Ref. 1). The coordinates along the abscissa are selected in such a way that Eq. (17) (phase mechanism of the limitation) corresponds to the straight lines 1 on the left- and right-hand sides of the figure. Curve 2 represents Eq. (18) for  $\chi''$  allowing for the nonlinear damping mechanism. The experimental points for the  $eph$  process on the right-hand side of this figure give the limiting values of the susceptibility in the region where low-frequency ( $\sim 100 \text{ kHz}$ ) oscillations of the absorbed microwave power are observed;  $p \equiv (h/h_c)^2$ .

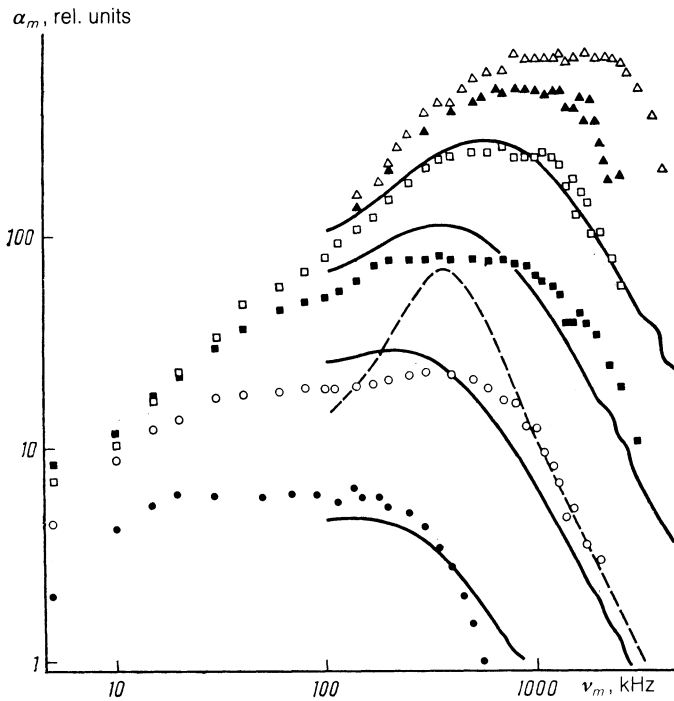


FIG. 5. Frequency dependences of the modulation response in the case when  $\nu_p = 9.33$  GHz,  $H = 0.75$  kOe,  $T = 1.8$  K, and the ratio representing the excess above the threshold is  $h/h_c = 1.12$  (●), 1.26 (○), 1.57 (■), 2.0 (□), 2.51 (▲), and 3.98 (△). The continuous curves are theoretical dependences for the case when  $|\eta/\mathcal{P}| \gtrsim 2$  and the dashed curve represents the theory in the case when  $|\eta/\mathcal{P}| = 0$ .

waves is a modulation method involving a study of the response of an excited system to a weak rf field which has practically no effect on the pumping threshold. This method has been used before to study the states beyond the threshold in the case of the  $nn$  and  $ee$  processes,<sup>9,10</sup> which makes it possible to estimate the contributions of the phase mechanism and of the nonlinear damping mechanism to the formation of a stationary state. We report the results of investigating the modulation response of a stationary state which appears as a result of the  $en$  pumping.

The points in Fig. 5 represent the experimental frequency dependence of the modulation response  $\alpha_m = \Delta P/H_m$  ( $\Delta P$  is the amplitude of oscillations of the absorbed microwave power), in the case of the  $en$  process obtained for different pumping rates. We can observe a characteristic linear rise at low modulation frequencies ( $\alpha_m \propto v_m$ ), followed by a fairly wide plateau and ending with a strong fall of the modulation response at high frequencies.

It should be pointed out that in the case of a smooth dependence  $\alpha_m(v_m)$  at frequencies  $\nu_s$  representing natural elastic vibrations of a sample there are singularities similar to those discussed in detail in Ref. 33 in connection with excitation of the  $nn$  process in antiferromagnetic  $\text{CsMnF}_3$ . As shown theoretically in Ref. 33, when the resonance frequency of collective oscillations of the parametric system is approximately  $\nu_s$ , i.e., when the condition for a collective acoustic resonance is satisfied,<sup>35</sup> such a system may become unstable because of the excitation of strong elastic vibrations the sample. If the parametric system has not yet reached the threshold of collective acoustic instability, then in the vicinity of the frequencies  $\nu_m \approx \nu_s$  the modulation response  $\alpha_m$  has a singularity where a narrow peak changes to a dip.<sup>33</sup> The amplitude of this singularity increases as the instability threshold is approached. The gain  $\alpha_m$  reached in Ref. 33 was a factor of  $\sim 10^2$ . Under the  $en$  pumping conditions the maximum gain was observed at  $\nu_m \approx \nu_s = 1.56$  MHz and amounted to 3. However, a collective-acoustic instability

was not observed in the  $en$  process. This was due to the fact that collective oscillations of the system of electron-nuclear pairs had a low  $Q$  factor, but the occurrence of a singularity of  $\alpha_m$  at frequencies  $\nu_m \approx \nu_s$  indicated coupling of the elastic vibrations of a sample to collective oscillations of the system of  $en$  pairs.

The experimental dependences of  $\alpha_m$  on the excess above the threshold are plotted in Fig. 6 for different modulation frequencies. When the excess above the threshold is small  $h/h_c - 1 \lesssim 1$ , the dependence of  $\alpha_m$  on  $h/h_c$  is strong as the frequency  $\nu_m$  is increased right up to  $\nu_m \sim \Gamma_e$ , but then the dependence begins to weaken.

We carry out a theoretical calculation of the modula-

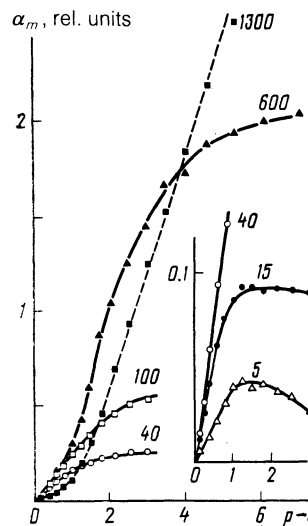


FIG. 6. Dependence of the modulation response on the excess above the threshold obtained for different modulation frequencies  $\nu_m$  [kHz]. The continuous curves are drawn through the points for the sake of clarity. The dashed curve is the theoretical dependence obtained for the case when  $|\eta/\mathcal{P}| \gtrsim 2$ ,  $\nu_m = 1300$  MHz, and  $p = (h/h_c)^2$ .

tion response  $\alpha_m$  using Eqs. (7) and (8), where the modulation field occurs in the parameter  $\Delta$  in the form

$$(\partial\omega_e/\partial H + \partial\omega_n/\partial H)H_m \cos(2\pi\nu_m t).$$

The parameter representing relaxation of electron spin waves will be assumed to depend on the number of nuclear magnons  $\gamma_e = \gamma_{e0} + \eta N_n$ , so as to ensure agreement with the susceptibility data  $\chi''$  (preceding section). It is assumed that forced oscillations of the medium near the equilibrium values of  $N_e, N_n$ , and  $\theta$  are small, so that the system (7)–(8) can be linearized in terms of the deviations from these values  $\delta N_e, \delta N_n$ , and  $\delta\theta$ . If we assume that

$$\delta N_e = A_1 \cos \omega_m t + B_1 \sin \omega_m t, \quad (21)$$

$$\delta N_n = A_2 \cos \omega_m t + B_2 \sin \omega_m t, \quad \delta\theta = A_3 \cos \omega_m t + B_3 \sin \omega_m t,$$

we find that the solution of the problem reduces to the solution of a linear system of the type

$$\begin{vmatrix} -\omega_m & \gamma & 0 & -R & 0 & S \\ \gamma & \omega_m & -R & 0 & S & 0 \\ 0 & -\gamma & -\omega_m & \gamma_n & 0 & S \\ -\gamma & 0 & \gamma_n & \omega_m & S & 0 \\ 0 & \tau_1 & 0 & \tau_2 & -\omega_m & G \\ \tau_1 & 0 & \tau_2 & 0 & G & \omega_m \end{vmatrix} \begin{vmatrix} A_1 \\ B_1 \\ A_2 \\ B_2 \\ A_3 \\ B_3 \end{vmatrix} = \begin{vmatrix} 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ A \end{vmatrix}, \quad (22)$$

where

$$\begin{aligned} \gamma &= \gamma_n N_n / N_e, & R &= \gamma_n - 2\eta N_e, \\ S &= -2\mathcal{F} N_e N_n, & \tau_1 &= 2\mathcal{F}_e + \mathcal{F}_{en} + \mathcal{F}(N_e + N_n) / 2N_e, \\ \tau_2 &= 2\mathcal{F}_n + \mathcal{F}_{en} + \mathcal{F}(N_e + N_n) / 2N_n, & G &= \gamma_n (1 + N_n / N_e), \\ A &= -(\partial\omega_e/\partial H + \partial\omega_n/\partial H)H_m. \end{aligned}$$

Now we have to find the relationship of  $\delta N_e, \delta N_n$ , and  $\delta\theta$  with the absorption signals at frequencies  $\omega_p \pm \omega_m$ , which are known<sup>36</sup> to be responsible for the depth of amplitude modulation of a carrier of frequency  $\omega_p$ . The amplitudes of these signals do in fact determine the experimental modulation response  $\delta_m$ .

A specific feature of our theory<sup>25</sup> is that of all the forms of motion of the magnetic moment of the crystal we select only the forced oscillations of combined magnon pairs of frequency  $\omega_p$ , which are excited parametrically by an external microwave field. The absorption spectrum of such a system is a line of frequency  $\omega_p$  which appears beyond the threshold of the resonance  $h > h_c$ . Application of a magnetic field of relatively low frequency ( $\omega_m \ll \omega_p$ ) results in magnetic modulation of the magnon spectrum, which (because of the nonlinearities of the parametric system) causes mixing of oscillations at the carrier frequency  $\omega_p$  with oscillations of frequency  $\omega_m$ . The result is manifested by the appearance of signals at side frequencies  $\omega_p \pm \omega_m, \omega_p \pm 2\omega_m$ , etc., which are additional to the main signal at the frequency  $\omega_p$ . The complete information on the absorption spectrum of such a parametric system is given by an expression for the power absorbed by a sample from two oscillating fields:

$$P(t) = -\partial\mathcal{H}/\partial t, \quad (23)$$

where  $\mathcal{H}$  is the reduced Hamiltonian of excited waves (see

Ref. 25) which allows also for the interaction of these waves with the modulation field. The explicit form of  $P$  is obtained from the familiar equations of motion<sup>25</sup> in the case of complex amplitudes of the excited waves (dissipation-free case). Then, transition to the "slow" variables  $N_e, N_n$ , and  $\theta$  (which in this case is analogous to averaging over the period of the pump field  $2\pi/\omega_p$ ) gives

$$P = 2\hbar\omega_p h V (N_e N_n)^{1/2} \cos \theta + \hbar\omega_m H_m \sin \omega_m t (N_e \partial\omega_e/\partial H + N_n \partial\omega_n/\partial H). \quad (24)$$

In the absence of the modulation field ( $H_m = 0$ ), this expression describes the absorption at the frequency  $\omega_p$ :

$$P_0 = 2\hbar\omega_p h V [N_e^{(0)} N_n^{(0)}]^{1/2} \cos \theta^{(0)}, \quad (24a)$$

where

$$N_n^{(0)} = \frac{\gamma_{e0}}{\eta} \left[ \left( \frac{h}{h_c} \right)^2 - 1 \right], \quad N_e^{(0)} = \frac{\gamma_n N_n^{(0)}}{\gamma_{e0} + \eta N_n^{(0)}},$$

$$\cos \theta^{(0)} = \{ \gamma_n [\gamma_{e0} + \eta N_n^{(0)}] \}^{1/2} / h V.$$

On application of a modulating field in Eq. (24), we obtain terms responsible for the intensity of absorption at the side frequencies separated from  $\omega_p$  by  $\pm \omega_m, \pm 2\omega_m$ , etc. In the case of a vanishingly small amplitude of the modulating field, we can simplify Eq. (24) by limiting it to terms linear in  $H_m$ . Then, we find that

$$P = P_0 + \delta P(t),$$

where

$$\begin{aligned} \delta P &= \Delta P \cos(\omega_m t + \varphi) = 2\hbar\omega_p h V \{ 1/2 \cos \theta^{(0)} \\ &\times [N_e^{(0)} N_n^{(0)}]^{-1/2} [N_n^{(0)} \delta N_e + N_e^{(0)} \delta N_n] \\ &- [N_e^{(0)} N_n^{(0)}]^{1/2} \delta\theta \sin \theta^{(0)} \} \\ &+ \hbar\omega_m H_m \sin \omega_m t \cdot [N_e^{(0)} \partial\omega_e/\partial H + N_n^{(0)} \partial\omega_n/\partial H] \end{aligned} \quad (25)$$

describes the intensity of absorption at the nearest side frequencies  $\omega_p \pm \omega_m$  of interest to us. Substituting in Eq. (25) the values of  $\delta N_e, \delta N_n$ , and  $\delta\theta$  calculated with the aid of Eqs. (21) and (22), we readily obtain  $\Delta P$  and finally the modulation response  $\alpha_m$ . It is found that the main contribution to  $\Delta P$  in Eq. (25) at frequencies  $\nu_m \lesssim 10$  MHz comes from the first term, which is proportional to  $\omega_p$ . The calculated results are presented in Figs. 5 and 6. Allowance for strong positive nonlinear damping of electron magnons (which makes it possible to describe the data for  $\chi''$  in Fig. 4) provides a satisfactory description of the modulation response of the *en* system (continuous curves in Fig. 5 and dashed curve in Fig. 6). For comparison, the dashed curves in Fig. 5 give the theoretical dependence  $\alpha_m(\nu_m)$  in the absence of nonlinear damping ( $\eta = 0$ ) for one experimental batch: a considerable discrepancy between the theory and experiment is then found to begin already at  $\nu_m \sim 1$  MHz. The linear fall of  $\alpha_m$  on reduction in  $\nu_m$  in low frequencies is not described by the theory given above, although this description can be provided phenomenologically by allowing for the drift of a packet of parametric magnons, as is done in the case of degenerate pumping.<sup>10</sup>

## CONCLUSIONS

We now summarize the main conclusions.

1. The validity of theoretical expression (2) for the threshold of a combined parametric resonance was confirmed experimentally.

2. Theoretical and experimental investigations were made of the influence of modulation of the spectrum of spin waves on the threshold of a combined  $en$  process. It is shown that at high modulation frequencies (high characteristic magnon relaxation frequency) the experimental results are described satisfactorily if we adopt the theory.

3. The hypothesis of strong positive nonlinear damping of electron magnons, due to a high nonequilibrium population of nuclear magnons, explains the absence of the "hard" excitation of the  $en$  process and also provides a satisfactory theoretical description of the susceptibility  $\chi''$  above the threshold and of the modulation response  $\alpha_m$ .

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