

Modulation method of investigating spin wave beyond the parametric-excitation threshold

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The behavior of nuclear spin waves (NSW) in the antiferromagnet CsMnF₃ above the threshold of their parametric excitation is investigated theoretically and experimentally under conditions when the magnetic field is modulated. Good agreement is obtained between the main conclusions of the theory and the experiment at above-threshold levels up to ~7 dB. The coefficient S_k of the nonlinear interaction of parametric NSW is calculated from the experimental data. A new method is proposed for the measurement of spin-wave relaxation; this method does not require absolute measurements of the amplitude of the microwave field at the sample. It is established that the NSW relaxation rates measured at and above the parametric-excitation threshold are equal.

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A considerable role in the investigation of the properties of magnetically ordered substances has been assumed recently by experiments on parametric excitation of spin waves (SW) by the method of parallel microwave pumping. The gist of the method is the following. The investigated sample is placed in a magnetic field $\mathbf{H} = \mathbf{H}_0 + \mathbf{h} \cos \omega_p t (\mathbf{h} \parallel \mathbf{H}_0)$. At a certain pump amplitude $h > h_c$ a parametric instability of the SW is produced in the system and is due to the decay of a microwave-pumping quantum into two magnons with wave vectors \mathbf{k} and $-\mathbf{k}$ and with energies $\hbar\omega_{\mathbf{k}} = \hbar\omega_{-\mathbf{k}} = \hbar\omega_p/2$. From the value of the threshold field hc it is possible to obtain information on the relaxation properties of the spin system as functions of the temperature and of the magnetic field. Equally interesting is the study of a system of parametric spin waves (PSW) above the threshold of their excitation. According to the theory developed by Zakharov, L'vov, and Starobinets (called the S -theory, see the review¹), the state of the PSW is characterized by two parameters, by the number N_k of the parametric waves and by the temporal phase Ψ_k of a pair of PSW with oppositely directed wave vectors. These parameters are interrelated, and this leads in particular to a phase mechanism that limits the number of the PSW. Experiments performed on electronic SW of yttrium iron garnet (YIG)^{2,3} and of an antiferromagnet with easy-plane anisotropy (AFEP)⁴ confirm the main conclusions of the S -theory. Further development of the theory was stimulated by investigations of PSW under conditions of modulation of the external magnetic field $\mathbf{H}_m \cos \omega_m t (\mathbf{H}_m \parallel \mathbf{H}_0)$. It was found⁵ that oscillations of the microwave power absorbed by the sample, at the modulation frequency ω_m , are observed directly above the parametric-excitation threshold. This phenomenon has so far not been observed, although it was used to record the threshold of parallel microwave pumping.^{6,7}

The present paper is devoted to a detailed theoretical and experimental investigation of the behavior of SW above the parametric-excitation threshold under conditions when the magnetic field is modulated, and in particular to an explanation of the very fact that modulation is induced by the microwave power absorbed above the threshold. The object

of the investigation was chosen to be nuclear spin waves (NSW) (see, e.g., Ref. 8) and AFEP, whose behavior above the parametric-excitation threshold has hardly been investigated heretofore. The validity of treating the above-threshold behavior of NSW within the framework of the S -theory is due to the fact that at helium temperatures these excitations have a clearly pronounced spatial dispersion; in this case the NSW spectrum forbids first-order decay processes (see Ref. 1). The measurements were performed on single crystals of CsMnF₃.

THEORY

In a modulated external magnetic field, the equations for N_k and $\theta_k = \pi/2 - \Psi_k$ are of the form^{1,7a}

$$\frac{d}{d\tau} \theta_k + b \sin \theta_k = a \cos \Omega_m \tau + r N_k + \Delta_k, \quad (1a)$$

$$N_k^{-1} \frac{d}{d\tau} N_k = -1 + b \cos \theta_k. \quad (1b)$$

We use here the notation

$$\tau = 2\gamma_k t, \quad b = \frac{h}{h_c}, \quad a = -\frac{\partial \omega_k}{\partial H_0} \frac{H_m}{\gamma_k},$$

$$r = -\frac{S_k}{\hbar \gamma_k \mathcal{N}}, \quad \Omega_m = \frac{\omega_m}{2\gamma_k}, \quad \Delta_k = \frac{1}{\gamma_k} \left(\frac{\omega_p}{2} - \omega_k - 2 \frac{T_k N_k}{\hbar \mathcal{N}} \right),$$

ω_k and γ_k are the frequency and damping decrement of the PSW; $h_c = (\hbar/g\mu_B)\gamma_k/V_k$ is the threshold pump amplitude; V_k is the coefficient of the coupling of the PSW with the pump field; \mathcal{N} is the number of magnetic ions in the sample; T_k and S_k are integral nonlinear-interaction coefficients responsible for the renormalization of the spectrum of the PSW and the pump. For simplicity, the terms customarily used in the S -theory

$$\sum_{\mathbf{q}} T_{\mathbf{kq}} n_{\mathbf{q}}, \quad \sum_{\mathbf{q}} S_{\mathbf{kq}} n_{\mathbf{q}}$$

are replaced in this case in the right-hand side of (1a) by $T_k N_k$ and $S_k N_k$.

It follows from (1) and (1b) that in the absence of a mo-

dulating field the principal parameters of the PSW system arrive at equilibrium values

$$\theta_k^{(0)} = \arccos(1/b), \quad N_k^{(0)} = (b/r) \sin \theta_k^{(0)}, \quad \Delta_k^{(0)} = 0. \quad (2)$$

When the field modulation is turned on, stimulated oscillations of N_k and θ_k set in near their equilibrium values (2). It must be noted that the oscillations of the number of PSW are due only to oscillations of the phase of a pair of PSW, in the equation for which the dependence of the field wave enters explicitly. In the simplest case, when the influence of the modulation of the pump threshold is small it suffices to use an approximation linear in H_m . It is then easy to obtain from Eqs. (1a) and (1b)

$$\theta_k - \theta_k^{(0)} = -\frac{\partial \omega_k}{\partial H_0} \frac{H_m}{\gamma_k} \cos \varphi \cos(\omega_m t + \varphi), \quad (3a)$$

$$\Delta N = N_k - N_k^{(0)} = -\frac{\partial \omega_k}{\partial H_0} \frac{2H_m \mathcal{N} \gamma_k}{S_k \omega_m} \left[\left(\frac{h}{h_c} \right)^2 - 1 \right] \cos \varphi \sin(\omega_m t + \varphi), \quad (3b)$$

where

$$\cos \varphi = 2\gamma_k \left[\left\{ \omega_m - \xi \frac{(2\gamma_k)^2}{\omega_m} \left[\left(\frac{h}{h_c} \right)^2 - 1 \right] \right\}^2 + (2\gamma_k)^2 \right]^{-1/2},$$

$$\xi = \frac{S_k}{S_k}$$

$$S_k = 2T_k + S_k,$$

Solutions (3a) and (3b) are valid at

$$H_m \ll H_m^* = \min \left\{ 1, \frac{S_k}{2\hbar\gamma_k} \frac{\omega_m}{\gamma_k} \left[\left(\frac{h}{h_c} \right)^2 - 1 \right]^{-1} \right\} \frac{\gamma_k}{|\partial \omega_k / \partial H_0| \cos \varphi}. \quad (4)$$

At $T = 2$ K, $\omega_m / 2\pi = 10^5$ Hz, and $(h/h_c)^2 = 7$ dB the estimate (4) yields $H_m^* \approx 1$ Oe.

It is of interest to note that with decreasing pump amplitude h , at a constant H_m , the amplitude of the stimulated oscillations of the number of PSW at the modulation frequency decreases and becomes equal to zero at $h = h_c$. This is the theoretical basis of the modulation method used in Refs. 6 and 7 to record hc. Owing to the oscillations ΔN_k , the microwave power absorbed at $h > h_c$ is amplitude-modulated

$$\Delta W = 4\gamma_k \hbar \omega_k \Delta N_k, \quad (5)$$

as was in fact observed in an investigation of electron and nuclear spin waves in the AFEP MnCO_3 and CsMnF_3 in Refs. 5-7.

Since we shall deal hereafter mainly with a system of nuclear PSW, we write down the amplitude (3b) of the oscillations of the number of the PSW for this concrete case:

$$\Delta N_k = -\frac{4\pi \hbar \Gamma_k}{S_k} \mathcal{N} \left(\frac{g\mu_B}{\hbar} \right)^4 \frac{H_0 H_\Delta^2 H_m}{\omega_{jk}^4} \frac{v_n^2}{v_k v_m} \left[\left(\frac{h}{h_c} \right)^2 - 1 \right] \times \left[\left\{ \frac{v_m}{2\Gamma_k} - \xi \frac{2\Gamma_k}{v_m} \left[\left(\frac{h}{h_c} \right)^2 - 1 \right] \right\}^2 + 1 \right]^{-1/2}. \quad (6)$$

Here

$$v_k = v_n \left[1 - \frac{H_\Delta^2}{H^2 + H_\Delta^2 + \alpha^2 k^2} \right]^{1/2}, \quad v_j \equiv \frac{\omega_j}{2\pi}, \quad \Gamma_k \equiv \frac{\gamma_k}{2\pi}.$$

In CsMnF_3 we have $v_n = 666$ MHz, $H_\Delta^2 = 6.4/T$ kOe², and $\alpha = 0.95 \times 10^{-5}$ kOe-cm.

EXPERIMENTAL RESULTS AND THEIR DISCUSSION

Parametric NSW waves in CsMnF_3 were excited at a pump frequency $\nu_p = \omega_p / 2\pi = 1$ GHz in the temperature range 1.5-4.2 K. The measurement procedure described in Ref. 6 was used, but we investigated here not the effect of the modulation on the value of the threshold, but the depth of the amplitude modulation of the microwave power above the threshold of the parametric instability. The microwave signal from the output of a helical resonator containing the sample was fed to a receiver that separated the low-frequency envelope of the signal, which was subsequently amplified and synchronously detected. The output signal A_m of the synchronous detector is proportional to the amplitude modulation (ΔW) of the microwave power, and this amplitude is in turn proportional to ΔN , see (5). Whenever it was necessary to know the absolute value of ΔN , the signal A_m was calibrated in units of the power ΔW , and the recalculated into ΔN by Eq. (5). The sensitivity of our measurement system turned out to be quite high: we could record the oscillation amplitude $\Delta N \approx 10^{11}$ of a number of PSW pairs. This corresponds to oscillations of the sample magnetic moment with an amplitude

$$\langle \Delta M \rangle = \frac{(g\mu_B)^2 H_0}{2\hbar\omega_k} \left(\frac{g\mu_B H_\Delta}{\hbar\omega_{jk}} \right)^2 \left(\frac{\omega_n}{\omega_{jk}} \right)^2 \Delta N_k \sim 10^9 \mu_B. \quad (7)$$

In those cases when maximum sensitivity was unnecessary, the synchronous detector was replaced by a linear one and the receiver by a crystal microwave detector.

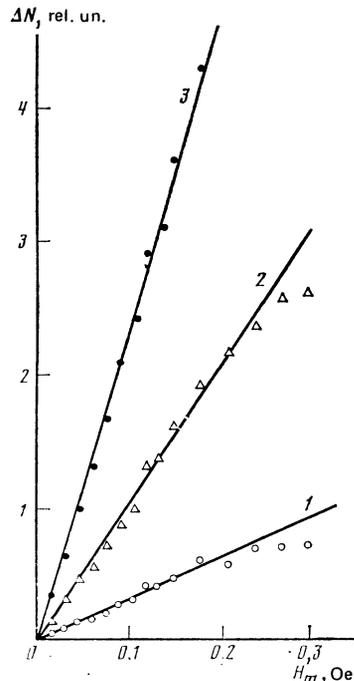


FIG. 1. Dependences of ΔN (of the amplitude of the oscillations of the number of PSW) on the amplitude H_m of the modulating field for several values of $(h/h_c)^2$: 1 - 1.9 dB, 2 - 4.2 dB; 3 - 7.6 dB $T = 1.96$ K, $\nu_m = 100$ kHz.

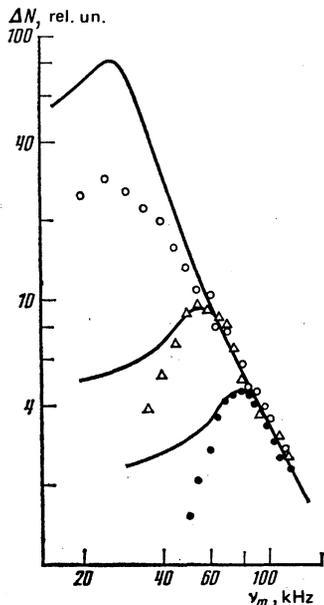


FIG. 2. Dependences of ΔN on the modulation frequency at various temperatures: \circ — 1.96 K; \triangle — 2.98 K; \bullet — 4.22 K. $(h/h_c)^2 = 6$ dB, $H_m = 0.04$ Oe. Solid lines—calculation by formula (6). The discrepancy between theory and experiment at low frequency is due to the influence of the modulation on the threshold h_c .

We verified first the functional dependences of ΔN which are defined by Eq. (6). Figure 1 shows the results of three sets of measurements of $\Delta N(H_m)$. It can be seen that they are described well by a linear relation $\Delta N \propto H_m$, in full accord with (6). Deviations from linearity set in at $H_m \gtrsim 0.25$ Oe, when the effect of the modulation on the parametric-pumping threshold comes already into play.

The theoretical dependence of ΔN on the modulation frequency ν_m is quite complicated. At $\nu m \gg \Gamma_k$, however, we should have $\Delta N \propto \nu_m^{-2}$. With decreasing ν_m the growth of ΔN slows down and the maximum is reached at $\nu_m \approx \xi \cdot 2\Gamma [(h/h_c)^2 - 1]^{1/2}$. This is precisely the $\Delta N(\nu_m)$ dependence observed in experiment (Fig. 2). This circumstance makes it possible to determine the PSW relaxation rate without absolute measurements of the microwave field h at the sample. It suffices for this purpose to measure the relative quantity h/h_c and find the position of the maximum on the $\Delta N(\nu_m)$ plot. The relaxation rate obtained in this manner agrees within the limits of error with the one determined from the parametric-resonance method, but the absolute measurement accuracy is higher in the proposed method. We note also that the equality of the relaxation parameters measured at and above the threshold of the parametric resonance (at $h/h_c \leq 7$ dB) indicates that at such an excess above threshold the relaxation is still independent of the number of the PSW.

It was very important to determine the limits of applicability of the theory with respect to the parameter $(h/h_c)^2$ that characterizes the ratio of the specified microwave power level to the threshold value. To this end, the NSW relaxation Γ_k was measured in a certain field, and then the $\Delta N(h/h_c)$ dependence. Knowing Γ_k and h/h_c it is possible to calculate from (6) the theoretical value ΔN^{theor} . Figure 3 shows a plot

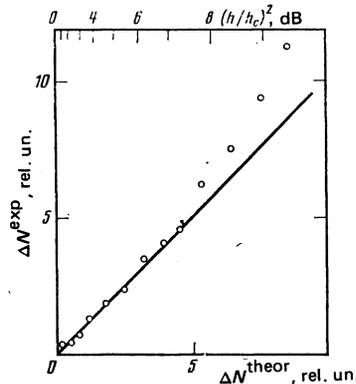


FIG. 3. Dependence of ΔN^{exp} on the excess above the threshold microwave power. $H_m = 0.09$ Oe, $\nu_m = 100$ kHz.

of ΔN^{exp} vs ΔN^{theor} . It can be seen that the theory describes the experimental results splendidly up to an excess ~ 7 dB above threshold.

We have thus succeeded in determining the range of the experimental values of H_m, ν_m , and h/h_c in which Eq. (6) describes adequately the functional dependences of ΔN on the indicated quantities (Figs. 1–3). It is undoubtedly of interest to compare the theory with experiment also quantitatively. The only parameter in (6) that does not lend itself to an exact numerical calculation is the integral pump renormalization coefficient \tilde{S}_k , since we do not know the angular distribution of the nuclear PSW. It is natural to assume on the basis of the theoretical calculation (given in the Appendix) of the nonlinear interaction coefficient \tilde{S}_{kq} that the substitution

$$\sum_q \tilde{S}_{kq} n_q = \tilde{S}_k N_k,$$

which was made for simplicity in (1a), reduces to the relation

$$S_k \approx (\Omega/4\pi) S_{kq}, \quad (8)$$

where Ω is the solid angle of the PSW distribution. The calculation of S_k under the simplest assumption that the production of the nuclear PSW is fully isotropic yields the following values of this coefficient:

$$S_k^{\text{theor}}(4.22 \text{ K}) = 2\pi\hbar \cdot 9.7 \text{ GHz},$$

$$S_k^{\text{theor}}(1.97 \text{ K}) = 2\pi\hbar \cdot 4.7 \text{ GHz}.$$

Using Eq. (6), we can determine experimentally the absolute value of the coefficient S_k and its dependence on H and T . To this end, the signal $\Delta W(H)$ was recorded with an automatic plotter at fixed values of h and T , see the curve of Fig. 4. From the parallel-pumping threshold we then determined the functions $\Gamma_k(H)$ and $h/h_c(H)$, and used Eq. (6) to calculate the coefficient S_k .

It was found that when the experimental values of $\Gamma_k(H)$ and $h/h_c(H)$ are substituted in (6) it is possible to describe an appreciable part of the $\Delta W(H)$ dependence under the assumption that S_k is constant, see Fig. 4 (the points indicate the calculated values of ΔW). The experimental values are $S_k^{\text{exp}}(4.22 \text{ K}) = 2\pi\hbar(1.14 \pm 0.3) \text{ GHz}$ and

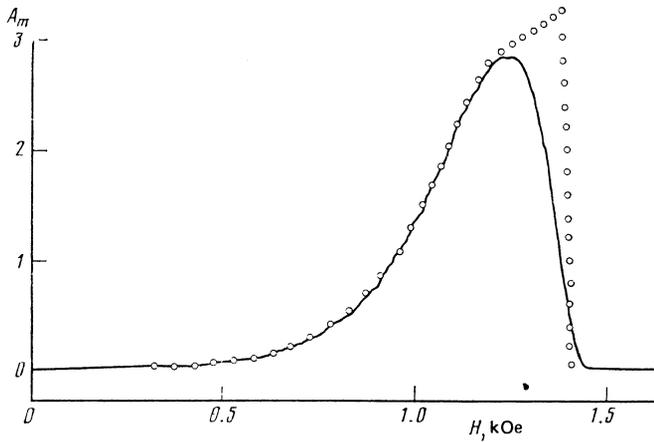


FIG. 4. Solid curve—automatically plotted signal $A_m \propto \Delta W$ at $T = 4.22$ K and $\nu_p = 1018$ MHz. The points correspond to calculation by Eq. (6) assuming $\tilde{S}_k = \text{const}$ (see the text).

$S_k^{\text{exp}}(1.97 \text{ K}) = 2\pi\hbar(0.2 \pm 0.05)$ GHz. They are not noticeably lower than the theoretical values obtained assuming full isotropy of the production of the parametric NSW. This points to a possibility of a strongly anisotropic PSW distribution that can be characterized by a parameter $\xi = \tilde{S}_k^{\text{exp}}/\tilde{S}_k^{\text{theor}}$ equal to $\xi(1.97 \text{ K}) \approx 0.04$ and $\xi(4.22 \text{ K}) \approx 0.12$. The assumption that the NSW have an anisotropic distribution is perfectly reasonable, since a strong anisotropy in the distribution of the electron PSW in CsMnF₃ was observed experimentally earlier in Ref. 9. There is as yet no quantitative explanation of so strong an anisotropy of the PSW distribution in an antiferromagnet.

On the other hand, the considerable difference between S_k^{theor} and S_k^{exp} can be due to the inadequacy of applying the model of Bose spin deviations to the description of NSW. In this case it is necessary to consider the modified S -theory directly within the framework of the spin representation.¹⁰

A discrepancy between theory and experiment (see Fig. 4) occurs in fields $H \gtrsim 1.25$ kOe, which correspond to the NSW wave-vector region $k \lesssim 0.7 \times 10^5 \text{ cm}^{-1}$. In this case, owing to the inhomogeneous broadening of the NSW spectrum the principal approximation $\Delta k/k \ll 1$ of the S -theory (Δk is the width of the NSW packet) ceases to hold.

As already noted in Ref. 6, it is convenient to use amplitude modulation of a microwave signal passing through a resonator with the sample to record the threshold of the PSW excitation. We estimated the accuracy of the registration of hc in our experiment by substituting in (6) $\Delta N \sim 10^{11}$, the experimental value of S_k , and other parameters. We obtain $h c^{\text{exp}} \approx h c \cdot 1.0005$, i.e., when $h c^2$ is exceeded by 0.1% (or 0.01 dB) the measurement system records the start of the parametric instability. This makes possible a substantial increase in the relative accuracy of the measurement of the SW relaxation at the parallel-pumping threshold.

We have investigated only the linear influence of the magnetic-field modulation on the parametric pumping of the spin waves. In this case the amplitudes of the oscillations of the number PSW, ΔN_k , and of the phase of a pair of PSW, $\Delta \theta_k$, about the quasi-equilibrium values are small, so that the stationary state of the PSW can be investigated. It turned

out that in this case all the experimental results can be described within the framework of the S -theory. With increasing h/h_c and H_m , however, the oscillations of N_k and θ_k cease to be small (in our experiments ΔN_k reached 10^{16} , i.e., $\Delta N_k \sim N_k$). An ever increasing role is assumed in the PSW system by nonlinear phenomena due to the modulation proper, and the linear theory no longer describes the observed experimental relations. An investigation of the behavior of PSW under these conditions is of independent interest.

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APPENDIX

The Hamiltonian that describes the scattering of NSW by one another

$$\mathcal{H}^{(4)} = \frac{1}{2N^2} \sum_{1,2,3,4} \Phi_{4n}(1,2;3,4) \alpha_1^+ \alpha_2^+ \alpha_3 \alpha_4 \Delta(\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3 - \mathbf{k}_4) \quad (\text{A.1})$$

can be obtained by using the results of Ref. 10 (α_k^+ and α_k are the NSW creation and annihilation operators). In non-symmetrized form, the interaction amplitude is

$$\Phi_{4n} = -\frac{J_0}{16\rho} [(3+\rho)(x_1+x_3) + 2(1-\rho)(x_1y_2+x_3y_4) + 8(1+\rho)x_1y_3 + 2(x_1y_3y_4+y_1y_2x_3) + 2x_1x_2y_3y_4], \quad (\text{A.2})$$

where

$$\rho^{-1} = \omega_n^2 / (\omega_1 \omega_2 \omega_3 \omega_4)^{1/2}, \quad x_k = (\omega_n / \omega_{jk})^2, \quad y = (g\mu_B H_\Delta / \hbar \omega_{jk})^2, \\ z = [\omega_{j_0}^2 + 6(g\mu_B H / \hbar)^2] / \omega_n^2.$$

Since Φ_{4n} depends, if the dipole-dipole interaction is neglected, only on the absolute values of the wave vectors of the interacting NSW, the coefficients $T_{\mathbf{k}\mathbf{q}}$ and $S_{\mathbf{k}\mathbf{q}}$ ($|\mathbf{k}| = |\mathbf{q}|$) can be represented in the form

$$T_{\mathbf{k}\mathbf{q}} = S_{\mathbf{k}\mathbf{q}} = -\frac{J_0}{4} \left(\frac{\omega_n}{\omega_k}\right)^2 \left(\frac{\omega_n}{\omega_{jk}}\right)^2 \left\{ \left[3 + \left(\frac{\omega_k}{\omega_n}\right)^2 \right] \left[\frac{1}{2} + \left(\frac{g\mu_B H_\Delta}{\hbar \omega_{jk}}\right)^2 \right] + \left(\frac{g\mu_B H_\Delta}{\hbar \omega_{jk}}\right)^4 \left[1 + \frac{1}{2} \left(\frac{g\mu_B}{\hbar \omega_{jk}}\right)^2 (7H^2 + H_\Delta^2 + HH_D) \right] \right\}. \quad (\text{A.3})$$

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