Instabilities of parametrically excited nuclear spin waves in antiferromagnetic $CsMnF_3$

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The behavior of a system of parametric nuclear spin waves in antiferromagnetic $CsMnF_3$ has been studied over a broad range of pump frequencies ($\nu_p = 700$–1050 MHz), over the temperature range $T = 1.7$–4.2 K, and over the supercriticality range $\xi = h/h_c - 1 = 0$–40, where h_c is the threshold for excitation of nuclear spin waves. Several instabilities of the steady state are observed when the frequencies of the nuclear spin waves ($\nu_k = \nu_p/2$) are in the range 350–450 MHz, where the parametric excitation of these waves is "hard". These instabilities develop in succession in the system with increasing ξ. A study of one of these instabilities shows that the nuclear subsystem does not affect the magnetic-field dependence of the velocity of sound waves with frequencies $\nu_s \ll \nu_k$.

In experiments on the parametric excitation of spin waves in magnetic materials by the method of parallel microwave pumping, a pump field $\mathbf{h} \cos \omega_p t (\mathbf{h} \| \mathbf{H}_0)$ is applied along with a static magnetic field H_0 to the sample. When the amplitude of this alternating field reaches a threshold $h = h_c$, an instability with respect to the decay of pump photons into pairs of magnons of half the frequency, with equal and oppositely directed wave vectors ($\nu_p = \nu_k + \nu_{-k}$) occurs. Above the threshold h_c, the number of parametric magnons increases exponentially with time until a balance is reached between the energy supplied to the system from the pump and the loss of energy due to the relaxation of the excited magnons. Above the threshold for the parametric process, the system is characterized by the number n_k of magnon pairs which are in resonance with the pump and by their temporal phase Ψ_k (Ref. 1).

The very first experiments on the parametric excitation of magnons in ferrimagnetic yttrium iron garnet revealed instabilities of the steady state taking the form of oscillations of the susceptibility above the threshold χ'' (Ref. 2). According to the theory for the steady state of magnons (the so-called S-theory[1]), this effect is due to a dipole-dipole interaction, which causes the amplitudes of the magnon-magnon processes to depend strongly on the directions of the wave vectors of the interacting magnons.[1] The situation is different in antiferromagnets, where the dipole-dipole anisotropy of the magnon-magnon interactions can be ignored.[3] In this case the S-theory predicts a spherically symmetric distribution of parametric magnons and the absence of instabilities of the steady state.

An instability of the steady state in a system of parametric electron magnons in an antiferromagnet was first observed in Ref. 4, where it was manifested as oscillations of the susceptibility of an $MnCO_3$ crystal above the threshold. Several instabilities were subsequently discovered in a variety of antiferromagnets,[5–11] but so far none has received an unambiguous theoretical explanation.

In the present paper we report a study of the behavior of a system of parametric nuclear spin waves in the easy-plane antiferromagnet $CsMnF_3$ at temperatures $T = 1.7$–4.2 K, at supercriticality levels $\xi = (h/h_c) - 1 = 0 - 40$, and at pump frequencies in the range $\nu_p = 700$–1050 MHz. The procedure for the parametric excitation of the nuclear spin waves and the characteristics of the crystal sample are described in Ref. 12. Here we simply note that the test samples, rectangular parallelepipeds with linear dimensions of 2–4 mm, are positioned at an antinode of the magnetic microwave field of a half-wave helical cavity filled with liquid helium, to avoid overheating the sample. With rare exceptions, the microwave pump is applied to the sample as a cw signal, and the spectrum of the signal at the resonator output is studied. Additional information can be found by using a modulation method to excite collective oscillations of the system of parametric nuclear spin waves.[13]

Most of the measurements were carried out in the range $\nu_p = 700$–900 MHz, where we have previously observed[12] "hard" excitation of nuclear spin waves in $CsMnF_3$, i.e., the existence of two threshold fields, h_{c1} and h_{c2} ($h_{c1} > h_{c2}$), the former corresponding to the appearance of parametric nuclear spin waves, and the latter to their disappearance. In this case we set the supercriticality ξ equal to the quantity $h/h_{c2} - 1$. In this frequency range we observe several distinct instabilities of the steady state of parametric nuclear spin waves, which we will discuss in the order in which they appear as the supercriticality is increased.

Before we discuss the results, we would like to call attention to a fundamental distinctive feature of experiments with nuclear spin waves. The mean free path of a nuclear magnon ($\sim 10^{-2}$ cm) is much smaller than the dimensions of the sample, so that the boundaries of the sample have a negligible effect on the behavior of the system of parametric nuclear spin waves, and all of these instabilities are undoubtedly three-dimensional. In work with electron magnons, in contrast, for which the mean free path is comparable to the dimensions of the sample, it is generally impossible to ignore the effects of surfaces.[11]

At pump frequencies $\nu_p \approx 800$ MHz, where the "hardness" of the excitation is most obvious, oscillations of the

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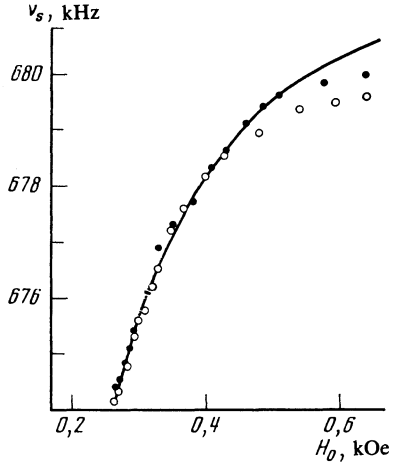

FIG. 1. Frequency of the most intense vibration mode of a parallelepiped sample with dimensions of $3.4\times2.9\times2.0$ mm versus H_0 at (○) $T=1.95$ K and (●) 4.2 K. The pump frequency is $\nu_p=802$ MHz. The solid line is a calculation with $\nu_{H_0\to\infty}$; $\omega_E\omega_{\rm med}/\gamma^2=1.6\cdot10^{-3}$ kOe^2.

pump power absorbed by the sample appear, even at the very lowest supercriticality levels. These oscillations are comparatively weak and are unstable in time. Their amplitude amounts to $\sim10^{-3}$ of the signal which is passed through the cavity (by way of comparison, the absorption level due to the parametric process is $\sim10^{-2}$). As h/h_{c2} is reduced, these oscillations disappear, precisely at $h=h_{c2}$ (within the measurement error $\approx0.5\%$), while as this ratio is increased to $h/h_{c2}=1.2$–1.7 the oscillations gradually convert into a stable harmonic signal with a frequency $F\approx2$ kHz, which depends slightly on T and H_0.[1)] With a further increase in ξ, this frequency increases, roughly in proportion to the supercriticality. The appearance of F is followed essentially immediately (the gap is <0.2 dB) by subharmonics $F/2$ and $F/4$. We also observe splitting of the fundamental frequency (or of the subharmonic) into a pair of closely spaced frequencies, F_1,F_2($F_1-F_2\approx0.2F$). When, as ξ is increased, the frequency F rises to the value corresponding to the relaxation rate of the nuclear spin waves under the given experimental conditions, the signal F generally disappears, while the subharmonic frequency continues to increase. The $F/2$ signal then goes through the same evolution. When a degree of supercriticality $\xi\approx2$–3 is reached, the spectrum of the signal becomes random.

It is noteworthy that this is the first observation of an instability of parametric spin waves in antiferromagnet supercriticality ($<5\cdot10^{-2}$). All of the instabilities which have been observed previously in a system of parametric electron magnons have generally appeared at higher supercriticality levels, $\xi>3$. Interestingly, at pump frequencies $\nu_p\gtrsim900$ MHz, where the excitation of the nuclear spin waves is not very hard, this instability is not observed.

It is possible that at frequencies $\nu_p=700$–900 MHz the phase mechanism which limits the number of spin waves beyond the threshold for the parametric process[1] is not the basic mechanism, and other limitation mechanisms capable of producing an instability at such low supercriticality levels must be taken into account.

The appearance of subharmonics with a succession of period doublings is of course one of the possible roads to chaos. This is a universal mechanism, observed in many systems.[14] In our experiments a system of parametric nuclear spin waves is seen explicitly to follow this route to chaos. We have previously observed the sequential appearance of subharmonics, up to $F/8$, in studying a double parametric resonance of electron magnons in the same crystal.[15]

Let us return to the experimental results. The random nature of the signal persists to $\xi=3$–4, beyond which the spectrum again changes radically. Satellites positioned symmetrically with respect to ν_p, separated from it by 0.3–1 MHz appear. The frequencies (ν_{si}) of these oscillations do not depend on the frequency of the microwave pump; they vary slowly as a function of the pump power (with ≈0.3 kHz), but they do depend on the dimensions of the sample. These frequencies fall in the region of natural elastic vibrations of the sample. The oscillation spectrum contains about ten lines of different intensities, the strongest at ≈680 kHz in a parallelepiped sample with dimensions of $3.4\times2.9\times2.0$ mm. The spectral width of these lines is exceedingly small, no greater than 5 Hz.

An analgous instability has been observed previously in a system of parametric electron magnons in the antiferromagnets $FeBO_3$ (Ref. 7) and $MnCO_3$ (Ref. 8). This instability has been interpreted as the decay of a spin wave into an oscillation of an elastic mode and a secondary spin wave. In our case, this type of instability is observed in a system of nuclear spin waves. The frequency of the oscillations which are excited depends on the magnetic field but not on the temperature (Fig. 1). In constructing the curve in Fig. 1 we set the supercriticality at the level which maximized the oscillation amplitude at the given H_0. In fields $H_0\gtrsim0.5$ kOe this maximum is not reached because another instability is excited (more on this below). It appears to be this circumstance which explains the discrepancy between the results at the two temperatures in Fig. 1 in fields $H_0\gtrsim0.5$ kOe. The dependence of the frequency of the elastic vibrations on the magnetic field stems from a corresponding dependence of the sound velocity ($\nu_{si}\propto v$). In our case, the frequency of the acoustic oscillations which are excited is well below the region where the spectra of the sound and the nuclear spin waves intersect. An expression for the sound velocity in this limiting case was derived in Ref. 4:

$$v=v_s[1-\omega_E\omega_{med}/(\omega_{10}^2-\omega_E\omega_N)]^{1/2};\qquad(1)$$

where v_s is the unperturbed sound velocity, $\omega_E=2\gamma H_E$ (H_E is the exchange field), $\omega_{\rm med}$ is the effective frequency of the dynamic magnetoelastic interaction, ω_N is the parameter of the hyperfine interaction,

$$\omega_{10}^2=\gamma^2H_0(H_0+H_D)+\omega_E(\omega_N+\omega_{med})$$

is the frequency of the antiferromagnetic resonance, γ is the magnetomechanical ratio, and H_D is the Dzyaloshinskiĭ field. An interesting aspect of expression (1) is that the presence of the nuclear subsystem has absolutely no effect on the functional dependence $v(H_0)$. After the expression for ω_{10} is substituted into (1), the term $\omega_E\omega_N$ disappears.

The physical explanation for this behavior of the

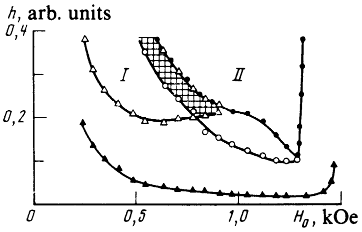

FIG. 2. I, Δ—Region in which the most intense vibration mode, with $\nu_s \approx 680$ kHz, exists; II—region in which harmonic oscillations of the absorption exist, for $T = 1.95$ K and $\nu_p = 802$ MHz. ▲) The threshold (h_{c2}) for the disappearance of the parametric nuclear spin waves; ● and ○) thresholds for the excitation and disappearance, respectively, of the oscillations. Which instability occurs in the hatched region depends on the direction in which h is varied. As h is increased out of region I, the vibrations ν_{si} persist, while as h is reduced from region II the harmonic oscillations persist.

phonon spectrum is as follows. As T is lowered, the hyperfine gap in the spectrum of the antiferromagnetic resonance becomes broader, and the NMR frequency decreases. As the NMR frequency decreases, the point at which the spectra of the sound and the nuclear spin waves intersect shifts to a low frequency, and it might seem that this shift would cause greater distortion of the spectrum of phonons with $\omega_s \ll \omega_n$. As the temperature is lowered, however, the point at which the phonons intersect the nuclear spin waves simultaneously rises, weakening the repulsion of the spectra. The theoretical results of Ref. 4 show that the net change in the sound velocity as a result of these two factors is strictly zero. The experimental results of the present study confirm that theoretical prediction. The behavior of the sound velocity as a function of the magnetic field (Fig. 1) is described by (1) with the value $\omega_E \omega_{\text{med}} / \gamma^2 = (1.6 \pm 0.2) \cdot 10^{-3}$ kOe^2. This value agrees well with the value estimated for ω_{med} from a measurement of the magnetoelastic constant.[16]

In a system of parametric spin waves, so-called collective oscillations may occur: oscillations in the number and temporal phase of magnon pairs with respect to their equilibrium values[1] (at the given ξ). These oscillations are excited in a linear way by the field $\mathbf{H}_m \cos \omega_m t (\omega_m \ll \omega_p, \mathbf{H}_m \| \mathbf{H}_0)$, and by varying the frequency ω_m we study their frequency spectrum, i.e., the frequency dependence of the intensity α_m of the collective oscillations. Because of the low quality factor of the collective oscillations, their spectrum has a rather broad maximum, at a position which depends on the relaxation rate of the parametric nuclear spin waves and on the supercriticality level.[1] In studying the spectrum we have found that even at the smallest values of ξ there are several narrow structural features against the background of the smooth spectrum, at frequencies ω_{si} corresponding to the natural acoustic modes of the sample (see the inset in Fig. 4). The existence of these structural features is evidence that the collective oscillations couple linearly with the sound, i.e., evidence of the existence of coupled collective-acoustic oscillations. These coupled oscillations were studied theoretically by Bakai and Sergeeva,[17] who introduced the concept of a collective-acoustic resonance, for which the condition is $\omega_s = \Omega_{\text{coll}}$ (Ω_{coll} is the resonant frequency of the collective oscillations). Since sound can be excited most easily at the frequencies ω_{si}, of natural elastic vibrations of the sample the quality factors of the coupled collective-acoustic oscillations are also highest at these frequencies. The relaxation rate of the elastic oscillations does not depend on ξ, while the damping rate of collective oscillations with frequencies ω_{si} falls off significantly with increasing ξ and as the system approaches the conditions for the collective-acoustic resonance. The result is an increase in the quality factor of the collective-acoustic oscillations with frequencies ω_{si}. At a certain threshold value of ξ, self-excited oscillations appear at frequencies ω_{si}. Because the spectrum of collective oscillations is wide, these self-excited oscillations exist not only at $\Omega_{\text{coll}}(\xi) = \omega_{si}$ (i.e., under these conditions of the collective-acoustic resonance) but over rather broad ranges of the distance above the threshold and the frequencies ω_{si}. The appearance of self-excited oscillations is seen experimentally as a modulation at frequencies ω_{si} of the microwave power which is transmitted through a resonator holding the sample. This interpretation of the instability also makes it a simple matter to explain the weak dependence of ω_{si} on the microwave power (see the discussion above). This dependence is evidently due to a change in Ω_{coll} and thus in the frequencies of the coupled collective-acoustic oscillations as ξ changes.

A different theoretical model was proposed in Refs. 8 and 18. That model explains the excitation of elastic vibrations in terms of the emission of a phonon by a magnon. The growth rate for that instability is determined by the total number of magnons with the given frequency and does not depend on how they are excited. That model apparently does not apply to our situation, since the phase correlation of the magnons plays an important role.

Yet another instability arises at approximately the same values, $\xi \approx 4$: strong harmonic oscillations of the absorption, at frequencies $\Omega = 2$–50 kHz (their strength is $\sim 10\%$ of that of the transmitted signal). The region (II) in the (h, H_0) plane in which this process occurs overlaps the region (I) in which the frequencies ν_{si} exist (Fig. 2). When the oscillations Ω appear, the signal at the frequencies ν_{si} is completely suppressed. The threshold at which the oscillations appear as ξ is increased is higher than the threshold at which they

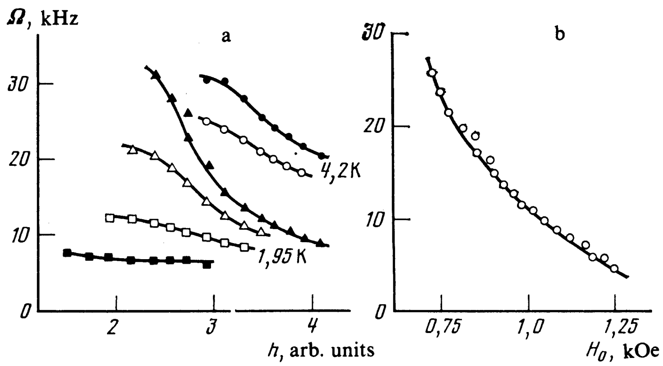

FIG. 3. a: The frequency (Ω) of the harmonic oscillations versus the amplitude (h) of the pump field for various values H_0, in kOe. ●,▲—0.75; ○,Δ—0.83; □—1.0; ■—1.17. b: The same frequency as a function of the magnetic field H_0 at the fixed value $h = 2.75$ (arbitrary units) and $T = 1.95$ K ($\nu_p = 700$ MHz).

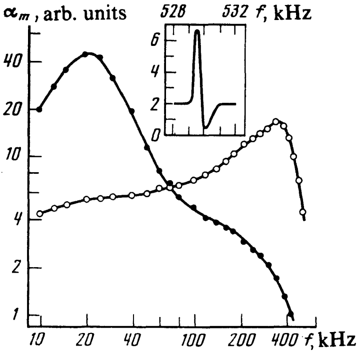

FIG. 4. Spectra of the collective oscillations of parametric nuclear spin waves for $\nu_p = 700$ MHz, $T = 4.2$ K, $H_0 = 0.7$ kOe, and $\xi = 5$ under conditions corresponding to the appearance of self-excited oscillations (open circles) and the appearance of harmonic oscillations (filled circles). The inset shows the spectrum of one of the collective-acoustic resonances.

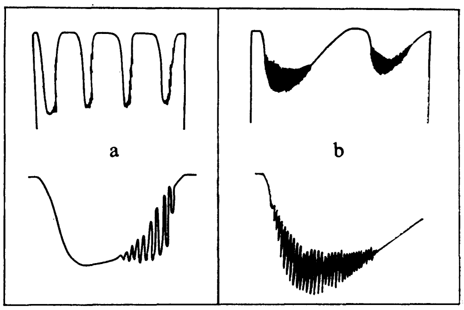

FIG. 5. Oscilloscope traces of the pulse at the exit from the resonator for $\xi = 20$, $\nu_p = 700$ MHz, $T = 1.95$ K, and (a) $H_0 = 0.4$ kOe or (b) $H_0 = 1.0$ kOe. The pulse length is 15 ms. The horizontal scale is enlarged by a factor of eight (a) or two (b) for the lower traces.

disappear as ξ is reduced; i.e., the excitation of the oscillations of this type is a hard excitation. In that region of H_0 in which both instabilities can occur, the particular instability which is observed in the hysteresis region (the hatched region in Fig. 2) depends on the direction in which h is varied. As h is increased out of region I, the vibrations with the frequency ν_{si} persist up to the threshold for the excitation of the oscillations, while as h is reduced the oscillations persist down to the threshold for their disappearance.

The most interesting aspect of these oscillations is the anomalous dependence of their frequency Ω on h. In contrast to all the instabilities which have been described previously, the frequency of the oscillations decreases with increasing h (Fig. 3a). At a fixed value of h, the frequency also decreases with increasing H_0 (Fig. 3b).

Figure 4 shows the frequency spectra of the collective oscillations, one of which was measured under conditions corresponding to the appearance of self-excited oscillations, while the other was measured during the excitation of strong harmonic oscillations Ω at the same supercriticality (the narrow structural features at the frequencies ν_{si} are not shown here). Interestingly, when the harmonic oscillations are excited there is a pronounced change in the structure of the spectrum. Its maximum shifts to low frequencies, while the intensity of the collective oscillations with the frequencies ν_{si} decreases dramatically; this behavior is apparently the reason for the disappearance of the self-excited oscillations. Further evidence for this interpretation comes from the fact that at pump frequencies $\nu_p \gtrsim 900$ MHz, where the excitation of strong harmonic oscillations Ω is not observed, the self-excited oscillations do not disappear anywhere up to the maximum supercriticality levels, $\xi \approx 30$.

As the supercriticality is increased to $\xi \gtrsim 20$, low-frequency oscillations (100–500 Hz) of a relaxation shape develop against the background of the harmonic oscillations, with a comparable intensity; their frequency depends weakly on H_0, T, and ξ. These oscillations were observed in two ranges of the magnetic field: near the boundary field (H_c) for the parametric process (for $\nu_p = 700$ MHz, and $T = 1.95$ K, this field is $H_c \approx 1.1$ kOe) and at $H_0 \approx 0.3$–0.5 kOe. The shapes and frequencies of the oscillations are different in these two regions (Fig. 5): The oscillation frequency near $H_0 = 0.4$ kOe is ≈ 500 Hz, while the oscillations observed near H_c have a frequency ≈ 100 Hz.

Similar relaxation oscillations were observed previously[9] in a study of parametric nuclear spin waves in $MnCO_3$. Those experiments were carried out at a high pump frequency, $\nu_p = 1170$ MHz, at $T = 1.25$ K. No instabilities of the steady state of the magnons were observed before the excitation of these oscillations. There is every reason to believe that these oscillations occur because the nuclear subsystem of the crystal becomes hot in comparison with the lattice. As a result of this overheating, the spectrum of nuclear spin waves changes, and the synchronization conditions which are required for the parametric process are violated. As a result, the transfer of energy from the pump to the nuclear subsystem decreases, and the nuclear subsystem cools off, over a time equal to the spin-lattice relaxation time; the process then repeats itself.

In those regions of H_0 where relaxation oscillations are not observed, the frequency of the harmonic oscillations continues to decrease with increasing ξ and the intensity of these oscillations simultaneously decreases. Interestingly, the lower boundary on the frequencies of these oscillations corresponds approximately to the relaxation rate of parametric nuclear spin waves. We recall that the same value is the upper bound on the frequency of the first of the instabilities discussed above.

In summary, at pump frequencies in the range $\nu_p = 2\nu_k = 700$–900 MHz, where we observe a hard excitation of nuclear spin waves in $CsMnF_3$, the state of the system of parametric nuclear spin waves is not steady at any degree of supercriticality, as is shown by various instabilities. In the absence of a suitable theory it is difficult to analyze these effects, but we hope that the results reported in this paper will stimulate the derivation of such a theory. It was suggested in Ref. 19 that the hardness effect is of a dislocation nature. We do not rule out the possibility that dislocations may also exert a significant effect on the formation of the steady state of parametric magnons.

We wish to thank V. L. Safonov for useful discussions.

[1] The hardness of the excitation in this case is $h_{c1}/h_{c2} - 1 \approx 0.5$.

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