Parametrically excited nuclear spin waves and magnetoelastic waves in antiferromagnetic $MnCO_2$ at high pumping amplitudes

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The behavior of a system of parametrically excited quasiparticles in antiferromagnetic $MnCO_3$ is investigated in the range of nuclear spin-wave frequencies. It is shown that the region of existence of "pure" parametric nuclear spin waves which is bounded from below on the (h, H) plane by the critical amplitude dependence $h_c(H)$ is bounded from above by superheat instability. The instability leads to finite overheating of the nuclear system relative to the lattice and to its transition into an inhomogeneous state. At a sufficiently high pumping amplitude, parametric excitation of magnetoelastic waves is observed in the region of excitation of nuclear spin waves with $k\rightarrow 0$. Their excitation is accompanied by generation, by the sample, of electromagnetic radiation at a frequency equal to half the pumping frequency, as well as by relaxation oscillations of the susceptibility. Transient processes and also the susceptibility and temperature of the nuclear system following parametric excitation of "pure" nuclear spin waves are investigated. It is shown that the predominant mechanism controlling the state of the system is the phase limitation of the magnon amplitude.

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1. NUCLEAR SPIN WAVES

Enough experience has by now been accumulated on the excitation of nuclear spin waves (NSW) by the parallel-pumping method. The result of this experience is that NSW can be easily excited in various types of antiferromagnets that contain the Mn^{2+} ion. These include cubic $RbMnF_3$ (Refs. 1 and 2), rhombohedral $MnCO_3$ (Refs. 3 and 4), and hexagonal $CsMnF_3$ (Ref. 1).

Nuclear spin waves are the result of indirect interaction of nuclear spins via an exchange-coupled system of magnetic moments of the ions.[5-9] The nuclear magnetic moment of an individual ion is coupled by way hyperfine interaction with the magnetic moment of the electron shell, which in turn is collectivized in a magnetically ordered system. The collectivized moments of the electron shell act via the hyperfine interaction on their own nuclear spins. Thus the nuclear spins turn out to be coupled in a spatial range corresponding to the propagation of a static perturbation in the magnetically ordered system. For an antiferromagnet, this range is

$$\lambda=[H_E^2/(2H_EH_A+H_0^2)]^{1/2}a,$$

where H_E is the effective exchange field, H_A is the anisotropy field, H_0 is the external magnetic field, and a is the lattice constant. As a result of this indirect interaction, the principal effective quantity is the nuclear magnetization $\langle m\rangle$ averaged over the range λ (over the volume λ^3), and collective oscillations of the nuclear magnetization become possible and can exert a strong influence on the spectrum of the magnetic system of the crystal. Since $(\lambda/a)^3$ is large, $\langle m\rangle$ averaged over the volume becomes a well-defined quantity, so that small deviations of its value can be considered and the equations can be linearized to determine the spectra of the natural frequencies in antiferromagnets with strong hyperfine interaction. Allowance for the hyperfine interaction leads to the following changes in the spectrum of the antiferromagnetic spin waves:

$$\Omega_{ik}^2=(\Omega_{ik}^0)^2+2\gamma_e^2H_EH_N, \tag{1}$$

Ω_{ik}^0 is the spectrum of the spin waves without allowance for the hyperfine interaction, H_N is the field acting on the electron system and produced by the nuclear magnetic system ($H_N\approx 10/T$ Oe, where T is the absolute temperature). Thus, an additional gap is produced in the antiferromagnetic spin-wave spectrum, proportional to the nuclear magnetization $\langle m\rangle$. In addition, a new branch of the spectrum is produced near the nuclear-resonance frequency (if the indirect interaction is disregarded)

$$\omega_{ik}^2=\omega_n^2(1-2\gamma_e^2H_EH_N/\Omega_{ik}^2). \tag{2}$$

In a number of antiferromagnets that contain the Mn^{2+} ion, the second term in the parentheses can be comparable with unity, i.e., the changes produced in the spectrum can be large. The oscillations corresponding to the spectrum (2) are the ones referred to as NSW. The spatial dispersion stems from the value of Ω_{ik} [Eq. (1)].

We investigate in this study magnesium carbonate $MnCO_3$. It is a rhombohedral crystal containing two magnetic manganese ions per unit cell. At a temperature 32.5 K, manganese carbonate turns into an antiferromagnet with weak ferromagnetism. The Mn^{2+} ion is in the S state and has a strong hyperfine interaction (ω_n = 640 MHz). The antiferromagnetic state has anisotropy of the easy-plane type (plane perpendicular to the S_6 axis). In the easy plane, there is practically no anisotropy. The static magnetic properties were investigated by Borovik-Romanov and Orlova[10] and by Borovik-Romanov.[11] Antiferromagnetic resonance was first observed by Date[12] and was investigated in detail by Borovik-Romanov, Kreĭnes, and Prozorova.[13] Nuclear magnetic resonance in the magnetically ordered state was observed by Borovik-Romanov and Tulin.[14] Parametric excitation of NSW in $MnCO_3$ was investigated by Yakubovskiĭ[3] and by Govorkov and Tulin[4,15]

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2. PROCEDURE

Parametrically excited NSW and their behavior beyond the excitation threshold were observed by studying the response of the susceptibility of the sample to the action of the pump field, both at the pump frequency itself and at the AFMR frequency. The exciting (and simultaneously measuring) cell constituted helical resonators for frequencies in the range from 1000 to 1200 MHz. The resonator was placed in a copper tube that served as a high-frequency shield as well as for mounting all the parts and the leads. The helix, several turns of copper wire (wire diameter 0.4 mm, turn diameter 5 mm) was placed in a foamed-polystyrene sleeve secured in the copper tube. The sleeve had an opening for a rod of 2 mm diameter and approximate length 14 mm. This rod served as a resonator at ~10 GHz for the observation of the AFMR. The sample was placed inside the helical resonator. The helix axis was parallel to the rod axis, so that the high-frequency magnetic field of the helical resonator (parallel to the helix axis) and of the "strip" resonator (perpendicular to the rod axis) were mutually perpendicular. The static magnetic field was parallel to the helix axis, so that the parallel-pumping geometry was realized for the natural oscillations of the helical resonator, and the AFMR-observation geometry for the "strip" resonator. The basal plane of the sample (easy plane) was horizontal.

A transmission measurement setup for both the decimeter band (~1000 MHz) and the centimeter band (10 GHz) was used. To measure small deviations of the signals passing through the cell, superheterodyne reception was used for both bands. The signal was observed directly either on an oscilloscope screen or with an automatic x-y reorder.

3. INSTABILITIES OBSERVED IN $MnCO_3$ SAMPLE AT A POWER HIGHER THAN CRITICAL FOR PARAMETRIC NSW EXCITATION

The purpose of this part of the study was to determine the boundaries of the region of existence of a simple system of parametrically excited NSW, and to study the instabilities that occur at these boundaries. The boundaries were determined by observing the dependence of the power passing through the helical resonator with the sample on the static magnetic field. The information was provided in this case by the breaks on the plot of the transmitted power against the magnetic field. In the course of operation, we tuned to maximum power transmission through the resonator, so that at small changes of the transmitted power the deviation from the constant level corresponded to the absorption in the sample, i.e., to the imaginary part of the susceptibility, $\Delta p(H) \sim \chi''(H)$. The measurements were performed in this case with the high-frequency field continuously acting on the sample.

Figure 1 shows typical plots of $\Delta p(H)$ for different irradiation power levels. There is no change in the susceptibility at low power levels. This is natural, since parametric excitation in parallel pumping is a nonlinear process and sets in at a certain threshold power. The 0.23 Oe and 0.8 Oe curves correspond to

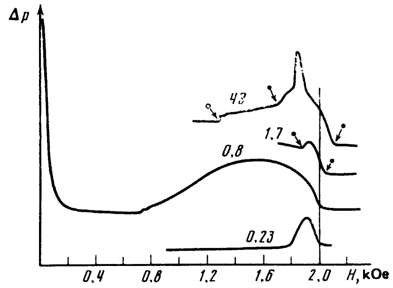

FIG. 1. Typical plots of the imaginary parts of the susceptibility of a sample under parallel pumping as a function of the magnetic field. The parameter on the curves is the amplitude of the pump field; f_p = 1117 MHz. T = 1.25 K.

the simple process of NSW excitation in parallel pumping in the entire range of existence of the additional absorption. The abrupt change of the power passing through the resonator in weak magnetic fields ($H < 0.2$ kOe) corresponds to a deviation of the resonator frequency because of the change of the active component of the sample susceptibility (of its antiferromagnetic system).

On the 1.7 Oe curve, in the region corresponding to nuclear resonance at the frequency $f_p/2$ [$H(k=0)$], additional absorption sets in above the background of the developed process of parametric NSW excitation. The evolution of this additional absorption with increasing irradiation power is much more complicated, as is seen from the 4 Oe curve. The vertical straight line near the 2.0 kOe field was drawn for the sake of clarity. It illustrates the fact that when only NSW are excited the absorption region does not extend into the stronger magnetic field, whereas after the secondary onset of the additional absorption it extends into the stronger magnetic field as the power is increased.

Naturally, in the presence of secondary additional absorption it is practically impossible to obtain data on the primary process from the sample susceptibility. On the other hand, however, it is of interest to determine the cause of this additional (secondary) absorption. We have investigated quite in detail the processes that limit the secondary additional absorption, both in strong and in weak magnetic fields. In a weak magnetic field, at sufficiently high power, the secondary additional absorption is produced jumpwise (the light circle in Fig. 1), and the jump amplitude increases rapidly as $H = 0$ is approached (with increasing irradiation power). In Fig. 2, the position of this jump on the (h, H) plane is marked by light circles.

To understand what it is that occurs at the instant after the susceptibility jump (corresponding to the branch 2), we tracked the position of the AFMR line on passing through the jump. The position of the AFMR line, as follows from Eq. (1), is determined not only by the observation frequency but also by the temperature of the nuclear system, or more accurately by its average magnetization $\langle m \rangle$. The method of determining the nuclear magnetization from the position of the AFMR line is somewhat limited by the fact that the observation is at one frequency (9.4 GHz in our case), which sets a

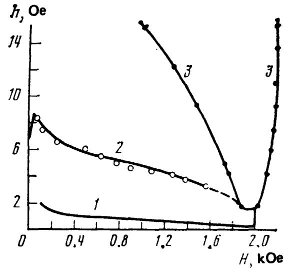

FIG. 2. Magnetic-field dependences of different critical fields in parallel pumping, obtained by reducing data similar to those in Fig. 1.

definite value of the magnetic field. However, by varying the frequency of the NSW pump and the temperature it is possible to study the process in different stages, both near the nuclear resonance field at the frequency $f_p/2$ ($k \to 0$) and far from this field. In the former case we can study the start of the process and in the latter its evolution during the later stages.

In Ref. 16 we described the results of observation of the influence of excitation of a nuclear spin system in the case of parallel pumping. It was shown there that the additional increase of the susceptibility, the limit of which is represented by curve 2 of Fig. 2, corresponds to a change of the nuclear temperature and of the wave vector of the NSW. The wave vector changes because of the change of temperature to satisfy the pumping condition, i.e., the pump frequency is equal to double the spin-wave frequency.

During the first stage, the average nuclear magnetization changes because of the excitation of the parametric NSW. This change is small but is of interest from the point of view of studying the degree of excitation of the parallel-pump system (this will be discussed somewhat later). This is followed by the onset of instability in the system, which leads to a change in the nuclear temperature. As seen from Eq. (2), at arbitrary nuclear temperature (or $\langle m\rangle$) the condition for

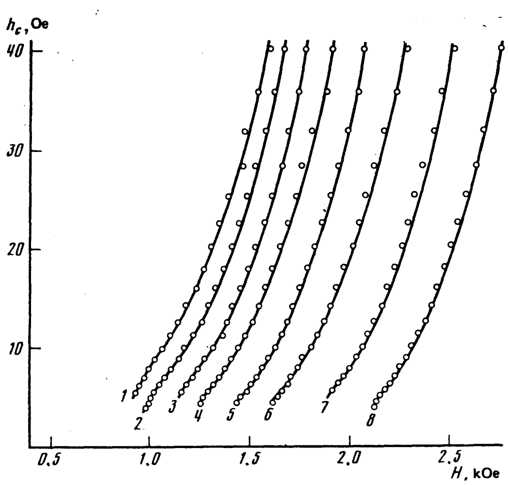

FIG. 3. Plots of the critical amplitude of the pump field for parametric excitation of magnetoelastic waves at different temperatures (for curves 1–8, respectively): 4.17, 3.6, 3.07, 2.68, 2.18, 1.8, 1.495, and 1.27 K. f_p=1100 MHz.

parametric excitation of NSW is satisfied by a certain set that form a line on the plane of $\langle m\rangle$ and k. It is along this line that the system of parametrically excited NSW builds up at the appropriate critical power. This power corresponds to line 2 of Fig. 2. With further increase of the power, the nuclear system is heated to a state at which $k \to 0$, and the nuclear temperature approaches a value corresponding to $\langle m\rangle_\omega$ [where $\langle m\rangle_\omega$ is the solution of Eq. (2) at $k \to 0$ and $\omega = 2\pi f_p/2$]. This corresponds to the maximum possible temperature of the nuclear spin system when heated through the NSW system (accurate to the line width of the homogeneous resonance). The presence of a broad AFMR line during the intermediate stage attests to an inhomogeneous spatial distribution of the average nuclear magnetization $\langle m\rangle$ (or of T_n). In addition, temporal oscillations of absorption are observed in AFMR, corresponding to temporal inhomogeneity of the magnetization $\langle m\rangle$.

4. SECOND THRESHOLD IN PARALLEL PUMPING AND GENERATION OF A SUBHARMONIC

We turn again to Fig. 1. It is seen there are when a certain critical pump amplitude (1.7 Oe curve) is reached, additional absorption is produced in the region of excitation of NSW with $k \to 0$. With increasing power, this absorption assumes a complicated character. On the weak-magnetic-field side, part of the wing of this absorption represents the onset of the superheat instability described above. The magnetic field that limits the additional absorption (dark circles on Fig. 1) is plotted as branch 3 in Fig. 2. As for the left-hand branch of this curve, it should be regarded as an assumption. The reason is that this branch is observed against the background of the variable temperature of the nuclear system and of magnetization oscillations that have already developed as a result of a NSW excitation. The complicated character of the absorption makes it difficult in this case to determine correctly the threshold amplitude of the alternating field.

More attractive is the right-hand branch of this curve. This branch extends into a stronger magnetic field relative to the nuclear-magnetic resonance field at the frequency $f_p/2$. In these fields there can be no excitation of NSW pairs because the energy conservation law $2\omega_k > \omega_p$ is violated. The process observed there takes place against the background of an unexcited nuclear system with specified external parameters and with a true value of the pump field. We have plotted in detail the dependence of the critical-field amplitude for the right-hand wing of this branch at different temperatures (Fig. 3). Unfortunately, the lack of corresponding calculations did not make it possible to reduce these data and obtain any fundamental quantities whatever.

The main circumstance that characterizes this process is the propagation of additional absorption towards the stronger magnetic field relative to the field of the excitation of NSW with $k \to 0$. Let us turn now to a qualitative treatment. Figure 4 shows the change of the spectrum of the nuclear magnons in one phonon branch of the crystal as a result of magnetoelastic interaction. That this excitation is real and that the sound velocity

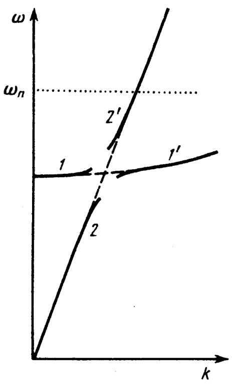

FIG. 4. Schematic representation of the change of the spectrum of the phonons and nuclear magnons in the region of magnetoelastic resonance. The dashed lines correspond to noninteracting branches of the phonons and nuclear magnons.

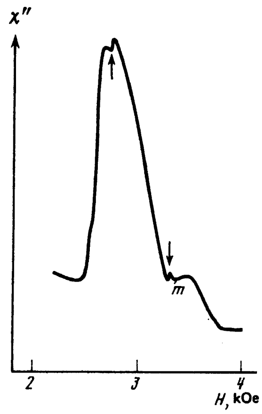

FIG. 5. Plot of the sample susceptibility for advanced oscillations.

changes near the magnetoacoustic nuclear resonance was demonstrated for MnCO by Gakel'.[17,18] Near the intersection, the spectra of the phonons and of the NSW become distorted by the magnetoelastic interaction. The wave vector that determines the intersection point is of the order of $k=\omega/s\approx 10^4\ \mathrm{cm}^{-1}$ (where s is the speed of sound) and is located in the region of inhomogeneous broadening of the NSW spectrum. Whereas the nuclear magnons are coupled to the exciting alternating field, the phonons do not interact with it directly in any way. A coupling with the alternating magnetic field does exist in the region of a sufficiently strong interaction due to the magnetic component of these waves. As the intersection point is approached, this connection becomes stronger and vice versa. This determines the steep character of the $h_c(H)$ plot. Propagation of the absorption towards stronger fields corresponds to excitation of the magnetoelastic wave from the lower part of the phonon branch on Fig. 4.

The form of the intersection of the spectrum singles out two magnetoelastic-wave sections with different properties. The section of the spectrum below the intersection with the nuclear-magnon branch is characterized by the fact that the heating of the nuclear magnetic system of the crystal upon excitation of the magnetoelastic waves leads to a rise of the spin-wave branch, to an increase of its distance to the exciting frequency, to a weakening of the coupling of the magnetoelastic waves with the pump field, and by the same token to a weakening of the heating. The sample introduces negative feedback into the heating of the sample by the pump power. Above the intersection with the magnon branch, on the other hand, the situation is reversed. Heating of the nuclear system leads here likewise to a rise of the spectrum of the nuclear magnons, but the coupling with the external field increases, thereby effectively increasing the pumping in the system and increasing the heating. In this region, the sample leads to positive feedback between the heating and the pump.

Figure 5 shows the variation of a signal passing through the resonator in the case of advanced excitation of magnetoelastic waves. Confirming the arguments advanced above, this figure demonstrates the strong asymmetry of the additional absorption. The absorption from the side of the strong magnetic field is much smaller and quite smooth. In a field weaker than $H(k=0)$, the absorption is considerably larger and has a number of additional peculiarities, on which it is advisable to dwell briefly. In a field $H(k=0)$ the absorption m has a minimum; it appears that this minimum separates the two described situations. Somewhat to the left of this minimum there is an absorption jump, marked by the downward arrow. This jump corresponds to the onset of oscillations in the absorption. A typical picture of these oscillations is shown in Fig. 6. They are of the relaxation type and fairly periodic. The period of the oscillation ranges from 2 to 6 msec. The chip at the peak (marked by the upward arrow in Fig. 5) corresponds to vanishing of the oscillations and to establishment of absorption in a position intermediate between the peak and the base of the oscillations. In this position, the signal contains a large high-frequency noise component.

To obtain some additional information on the situation in the case of excited magnetoelastic waves, we have attempted to detect the electromagnetic-field radiation from the sample.[19] On the whole it is quite difficult to expect radiation from a sample parametrically excited at frequencies different from that of the pump. This is possible in the case of excitation of uniformly precessing magnons or in the presence of a mechanism that converts the excited quasiparticles into homogeneous oscillations. Otherwise the averagings over the spatially inhomogeneous oscillation decrease the expected generation to unobservable values.

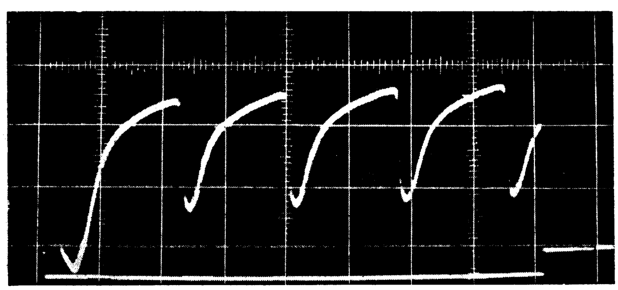

FIG. 6. Typical form of periodic oscillations.

To pick up the alternating magnetic field from the sample we placed alongside the helical resonator a turn connected in a resonant circuit having a natural frequency tunable between 500 and 700 MHz. The turn was made to approach from below, and its plane was vertical and parallel to the axis of the helix. The resonant circuit was connected to the receiver through a coaxial line.

When only NSW are excited, generation takes place near the position of the NSW with $k \to 0$. In the case of excitation of magnetoelastic waves the generation is observed in the entire range of their excitation. The generation is nonstationary and is deeply modulated by noise with characteristic frequencies 10–100 Hz. The power generated in this case is quite high. At such a poor reception (low filling factor) it reaches a value of the order of 10^{-8} W at a pump power 10^{-1} W. In the case of high pump power, generation was observed also in the region of existence of superheat instability. The generation has a rather narrow spectral band. We had no spectrum analyzer, and can present only a rough estimate. In the absence of oscillations and of high-frequency noise, this band is narrower than 10 kHz. In the presence of oscillation and especially in a state with high-frequency noise, the spectrum broadens to 10 MHz, with a rather complicated dependence of the amplitude on the frequency.

It appears that the generation of the subharmonic is observed whenever quasiparticles with wave vector $k < 5 \cdot 10^4$ cm^{-1} are parametrically excited. In the case of NSW this is realized within the limits of the nuclear-magnetic-resonance line width. Magnetoelastic waves also have a wave vector of the order $2\pi f_p/s \approx 10^4$ cm^{-1}. In the case of superheat instability the wave vector is likewise of the same order.

The opinion that the line width of AFMR (and of NMR in the case of strong dynamic coupling) is determined by the inhomogeneities of the crystal was advanced 15 years ago. By starting from the width, it is possible to obtain the characteristic wave vector of the inhomogeneity. For $MnCO_3$ it amounts precisely to the already mentioned several times 10^{+4} cm^{-1}. As shown by Eastman,[20] for $RbMnF_3$ this inhomogeneity is due to the presence of inhomogeneous deformation in the sample, determined by the defect structure of the crystal. It appears that this is also the reason for the inhomogeneities observed in a number of other crystals (including $MnCO_3$). It can therefore be suggested that the transformation of quasiparticles with wave vectors in this band into homogeneous oscillations is effected by inhomogeneities connected with the structure defects.

Besides the very presence of subharmonic generation, this fact indicates that the second threshold corresponds to generation of a pair of quasiparticles with identical energies, and in this band, other than nuclear spin waves, these can be only magnetoelastic waves. There is apparently no real splitting of the spectra as a result of the interaction in this case. What does occur is magnetoacoustic resonance, usually characterized by a strong increase of the quasiparticle relaxation rate in the crossing region. This can serve as a good model for the explanation of the susceptibility oscillations.

When a nuclear system is heated by excitation of magnetoelastic waves, a change in the state of these quasiparticles takes place. The wave vector of the quasiparticle changes in accord with the change of the nuclear temperature. In the unstable situations of interest to us at present, this wave vector decreases. An abrupt singularity occurring in the quasiparticle relaxation rate in the course of this decrease can stop the parametric excitation because of the increase of the critical field. The heating of the nuclear system is determined by the rate of quasiparticle generation, i.e., by the excess of the pump over the threshold. The cooling process is determined by the spin-lattice relaxation time T_1. The presence of these two times and of a singularity in the relaxation rate in the path of the system lead to oscillations of the susceptibility, and to a subsequent suppression and resumption of the quasiparticle generation. The same can be stated concerning the behavior of an NSW system following superheat instability. The role of the singularity is played as before by the location of the intersection of the spectra. In the first case, however, we approach the interaction point along the phonon branch in Fig. 4, and in the other along the magnon line (in fact, on the contrary, this point approaches the given value of the magnetic field and the frequency $f_p/2$). When observing the critical field of the parametric excitation, this singularity may not appear because of the already described inhomogeneous broadening of the sample spectra.

5. PARAMETRICALLY EXCITED NSW IN THE CASE OF STRONG PUMPING

Besides the descriptions of several independent phenomena, we have clarified in the preceding section the region of existence, in our system, of completely unencumbered parametrically excited NSW. This region (Fig. 2) is bounded from below by the critical amplitude of the excitation (branch 1) and from above by the amplitude at which the superheat instability sets in (branch 2). In the case of weak magnetic fields, it is necessary to exceed 0.5 kOe to avoid a noticeable inhomogeneity of the sample magnetization, whereas in strong fields the upper limit (for our sample) should correspond to excitation of NSW with wave vector $k \approx 5 \cdot 10^4$ cm^{-1} in order not to land in the region of possible excitation of magnetoelastic waves and inhomogeneous broadening, where the wave vector does not have a definite value. Within these limits it is possible to investigate a system of parametric NSW up to a ratio $h/h_c \approx 6$, (where h_c is the critical amplitude of the alternating pump field for the excitation of NSW).

We call attention first to the establishment of the stationary state in a system of parametrically excited NSW when the pump field is turned on. Figure 7 shows the inverted peak of the pulse that modulates the pump radiation (the larger the absorption the higher the curve), i.e., the ordinate is proportional to the imaginary part χ'' of the susceptibility). At a small ratio h/h_c (1.58) the increase of the susceptibility is monotonic and the stationary state is reached after a rather long time. During the initial growth stages the susceptibility is proportional to the amplitude of the oscillations of

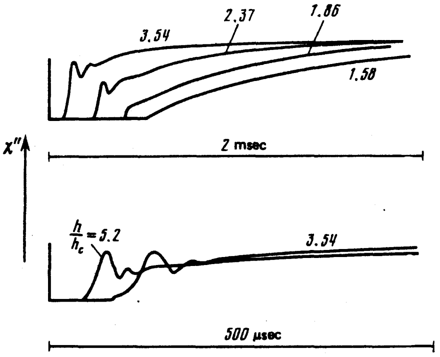

FIG. 7. Chip on the video pulse corresponding to excitation of nuclear spin waves in parallel pumping. The parameter marked on the curves is the supercriticality h/h_c.

the $\langle m\rangle_z$ component of the magnetization, i.e., to the number of the excited NSW. Therefore some important information is provided by the instant of time, after the start of the pulse, at which a noticeable change in the signal sets in. The sensitivity to this change is determined by the receiver, and does not change when two attenuators are used. This means that the start of the deviation corresponds to excitation of relatively equal number of NSW, and this number is determined by the sensitivity of the system (this quantity was not calibrated in terms of the number of the NSW).

The experimental plots of the time τ from the start of the pulse to the chip are well described by the relation

$$1/\tau \sim h/h_c - 1, \tag{3}$$

from which it can be found that a constant number of excited NSW corresponds to

$$\tau(h/h_c - 1) = \text{const}, \tag{4}$$

and this means that the increase in the number of NSW is determined by a certain function of the quantity $t(h/h_c - 1)$. Smirnov[21] has shown that for antiferromagnetic spin waves this functions is an exponential. For NSW, one can also expect an exponential with argument $t(h/h_c - 1)$, although we have not verified this.

We return now to Fig. 7. It is seen that at large ratio h/h_c the time dependence of χ'' becomes nonmonotonic. Damped oscillations with a period on the order of 30 μsec are observed. Figure 8 shows the dependence of the susceptibility in the steady state on the value of h/h_c. This dependence can be obtained both for long modulating pulses (~20 msec) and by the continuous technique (from data similar to Fig. 1). The $\chi''(h)$ curve as a characteristic form for the indicated magnetic-field range (0.5 kOe $< H < H(k=0) - \Delta H$). For nuclear spin waves excited near the position of the NMR line at the frequency $f_p/2$, the $\chi''(h)$ curve is more stretched out, i.e., the distance of the maximum from the origin is not 5 dB, as for the data of Fig. 8, but much larger. Damped oscillations appear on the susceptibility evolution curve at h/h_c corresponding to the maximum of the susceptibility $\chi''(h)$.

Thus, the behavior of the system of parametrically excited NSW exhibits the following characteristic fea-

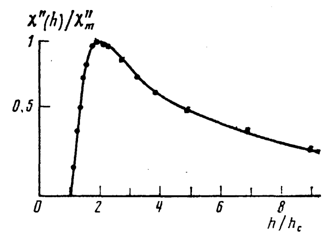

FIG. 8. Dependence of the imaginary part of the susceptibility of parametrically excited NSW in the stationary state on the supercriticality.

tures. The susceptibility of the system in the stationary state is nonmonotonic and can be represented by a perfectly concrete relation, plotted in Fig. 8 for a wide magnetic-field range. The establishment of the stationary state at h/h_c larger than 1.86 is characterized by damped oscillations of the susceptibility. The evolution of the parametric excitation during the initial stage can be described as a function of the argument $t(h/h_c - 1)$.

6. MECHANISM THAT LIMITS THE NUMBER OF SPIN WAVES IN PARAMETRIC EXCITATION

From among the foregoing results on the behavior of a system of parametrically excited NSW, the most significant is the dependence of the susceptibility on the amplitude on the pump magnetic field. This dependence can cast light on the main interaction that controls the number of the NSW. This state of a magnetically ordered crystal is characterized by an increased content of a certain group of spin waves, which interact in best fashion with the pump field. This increased spin-wave content determines the properties of the sample in the stationary state. Various interactions in the crystal alter a number of its properties. Since the number of spin waves determines the average magnetization in our case of a nuclear magnetic system, and the average magnetization governs the frequency of the spin waves, a change takes place in the spin-wave spectrum. The frequency change should not affect the behavior of the stationary state, since it leads to excitation of other spin waves that are reasonantly coupled with the external pump field. In the transition from the unexcited to the excited states, however, the change of the frequency can play a noticeable role. Depending on the ratio of the growth rate of the number of spin waves, which is a function of the amplitude of the pump field (3), (4), to the rate of return of the average nuclear magnetization to the equilibrium value, which is a constant of the material in first-order approximation, the transition can be either monotonic or oscillatory. The number of spin waves influences also the rate of the NSW relaxation, and this can already limit the number of the excited NSW if the increase of the number of spin waves leads increases the relaxation rate. Then, at a certain pump field amplitude exceeding the critical value, the number of spin waves decreases to a value at which the critical amplitude, which is directly connected with the relaxation rate, becomes equal to the amplitude of the external field. The dependence of the number of spin waves on the magnetic-field amplitude then takes on the meaning of the dependence of the relaxation rate on the number of spin waves, and this is already fundamental characteristic of the system. This so-called nonlinear-damping mechanism is described by a system susceptibility that increases monotonically with the pumpfield amplitude, something which does not occur for NSW in $MnCO_3$ (Fig. 8).

The number of spin waves can also be limited by the properties of the apparatus, more accurately, of the absorbing cell. If the Q of the absorbing cell is large, the onset of nonlinear absorption can decrease the amplitude of the pump field in the cell to the critical value. The $\chi''(h)$ dependence is then quantitatively determined by the Q of the resonator, and has qualitatively a form similar to that in Fig. 8, i.e., it first increases rapidly and then decreases monotonically like h^{-1}. In the investigations of NSW, the change of the signal following the excitation in our absorbing cell was very small ($\sim 10^{-2}$). This did not enable us to consider seriously the mechanism connected with the change of the Q of the absorbing cell.

One other important consequence of the presence of parametrically excited NSW in samples in the case of parallel pumping is the presence of an alternating magnetic field. This field is the result of the fact that the excitation of the spin waves is connected with the motion of the corresponding magnetization. It is this which leads, in accordance with the electrodynamics equations, to alternating magnetic fields. In the case of parallel pumping, the optimal conditions of coupling with the external field cause the phase of the spin waves to be constant with a good degree of accuracy (provided that the radiation wavelength $\lambda \gg d$, i.e., exceeds the sample dimensions). Constancy of the phase makes magnetic fields due to magnetization motion macroscopic. The existence of this effect has been known for a long time, and its consequence is the doubling of the radiation frequency at resonance in magnetically ordered crystals. This alternating magnetic field combines with the field of the wave and the resultant field leads to excitation

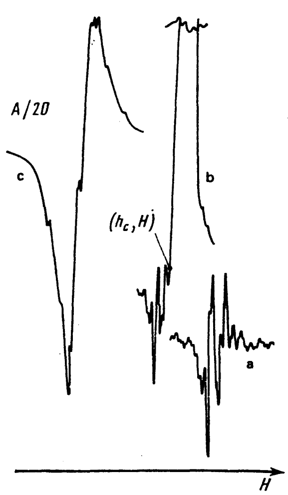

FIG. 9. Field dependence of signal proportional to the change of the position of the AFMR line when the NSW system is switched over from the state of parametric excitation to the state of equilibrium with the lattice. The pump amplitude increases from a to c.

of the spin waves in the sample and determines the stationary state of the system of parametrically excited NSW.

A theoretical analysis of the phase limitation mechanism was carried out by Zakharov, L'vov, and Starobinets,[22] who have shown that allowance for the self-action of parametrically excited systems weakens its coupling with the pump field and leads to a dependence of the susceptibility on the pump-field amplitude in a form corresponding to that shown in Fig. 8. In the real situation, of course, all the mechanisms that limit the growth in the number of spin waves act simultaneously, and one can speak of a dominant mechanism. In the investigated situations for antiferromagnets, for the case of both antiferromagnetic waves[23] and NSW,[4] the dominant mechanism is the phase limitation mechanism connected with allowance for the self-action of the parametrically excited system.

7. MEASUREMENT OF THE NUMBER OF EXCITED NSW IN PARALLEL PUMPING

A rather important role in determining the behavior of a system of parametrically excited NSW is played by the relation between the number of spin waves and the pump amplitude. As already mentioned, in the case when the number of spin waves is limited by nonlinear damping, this relation has the meaning of the dependence of the relaxation rate of the spin waves on their number. In the case of the phase mechanism, the theory yields the following relation between the number of spin waves and the pump-field amplitude[22]:

$$n^2 \sim (h/h_c)^2 - 1. \qquad (5)$$

In investigations of NSW, their number can be determined from the change of the nuclear magnetization, which can be measured indirectly by starting from the position of the antiferromagnetic-resonance line. In this case one measures not the number of parametrically excited spin waves, but the total change in the number of NSW. This total change consists of parametrically excited NSW and of NSW that result from relaxation of parametric spin waves. Inasmuch as in this case the cause of the change in the number of NSW is parametric excitation at constant nuclear spin-lattice relaxation, the total number of the NSW is proportional to the number of parametrically excited spin waves.

The measurement was by a two-frequency method. The parametric spin waves were excited in the usual manner by parallel pumping[4] and the AFMR line was simultaneously observed. To increase the sensitivity, the pumped power was modulated by pulses of long duration, so that the system was in the stationary state during most of the pulse. We used a meander at a frequency of 20 Hz. The antiferromagnetic resonance was observed with a superheterodyne receiver whose output signal was fed to a narrow-band amplifier. The low-frequency signal was phase-detected and an x-y recorder plotted a curve proportional to the change of the AFMR line position. Parametric excitation of NSW caused a change in the nuclear magnetization at the modulation frequency. This led to a small shift of the

AFMR line also at the modulation frequency. The shift of the AFMR line caused a change of the signal in the microwave circuit. The signal was proportional to the quantity

$$(\partial\chi''/\partial H)\Delta\langle m\rangle, \quad (6)$$

where χ'' is the imaginary part of the susceptibility, corresponding to absorption in AFMR.

Figure 9 shows the resultant plots of signals of this type. Figure 9(a) shows the signal that results from the action on the nuclear system at irradiation amplitudes lower than critical. As seen from this curve, a change in the position of the AFMR in synchronism with the modulation of the pump power is observed even here. In Fig. 9(b), the pump power was chosen such that the critical amplitude was reached at the instant of maximum absorption for AFMR. It is seen that the left wing of the signal is the below-threshold influence, whereas the right wing increased rapidly in intensity and corresponds to the above-threshold influence. Figure 9(c) corresponds to the case when the parametric excitation takes place in the entire region of observation of AFMR.

Since the amplitude of the signal in Fig. 9(c) is proportional to $\Delta\langle m\rangle$, and $\Delta\langle m\rangle$ is proportional to the number of NSW, we can plot a quantity proportional to the number of spin waves as a function of the pump-field amplitude. Figure 10 shows for different frequencies the dependence of the square of the signal amplitude on the square of the external-field amplitude [in accord with Eq. (5)]. As seen from this figure, the results of the experiments are well described by straight-line segments. The kink corresponds to an increase of the slope of the line with increasing pump-field amplitude. These results show that the observed data can be described as the evolution of two successive processes of parametric excitation with somewhat different thresholds. The theory of phase limitation of the number of quasiparticles in parametric excitation states that the internal alternating field in the sample is the sum of the external field and of the field of the reaction of the sample; this combined field is equal to the critical field and is constant in the range of action of the phase mechanism.

The behavior of the high-frequency susceptibility

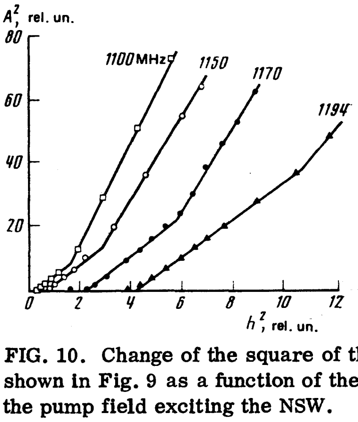

FIG. 10. Change of the square of the amplitude of the signal shown in Fig. 9 as a function of the square of the amplitude of the pump field exciting the NSW.

of the sample in the case of parallel pumping, as well as other data, offers convincing evidence that the phase mechanism of limitation of the number of spin waves acts in the NSW system. Ozhogin *et al.*[24] have shown that phase relations play a most important role in parallel pumping in an NSW system. The theory of Zakharov, L'vov, and Starobinets[22] posulates constancy of the internal alternating field, while the results show that in the range of action of the phase mechanism one observes one other threshold process of approximately the same power $[\partial(N^2)/\partial(h^2)]$ as the first. These results are easiest to reconcile by introducing nonlinear damping. When the parametric excitation influences differently the damping of the different NSW groups, the critical field for certain groups can coincide, as a result of which another group of nuclear magnons becomes excited.

The characteristic number N that can be excited in parallel pumping is 10^{18} cm^{-3}. Of course, not all these NSW are parametrically excited. There is also a large number of nuclear magnons that are not in equilibrium and are the results of the decay and re-excitation in relaxation processes, which also lead to a decrease of the average nuclear magnetization.

CONCLUSION

We have investigated the behavior of the susceptibility of antiferromagnetic $MnCO_3$ under parallel pumping in the NSW excitation range. We investigated the processes that limit the region of existence of "pure" NSW. We have shown that this region is bounded on the high-pump side by an instability that manifests itself in the form of superheating of the nuclear magnetic system. It may be caused by the instability of the phase mechanism when the pump exceeds greatly the critical value.

We studied the state of the system after the onset of a second threshold in the NSW region with wave vector $k \to 0$. We have shown that the above-threshold behavior agrees with the assumption that magnetoelastic waves are excited in this process. Strong above-threshold oscillations and generation of a subharmonic are observed.

We have investigated the behavior of a system of parametrically excited NSW at the possible values of the supercriticality. We have shown that the dominant mechanism determining the stationary state of the system is the phase mechanism of limitation, proposed by Zakharov, L'vov, and Starobinets.[22]

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