Investigation of nonlinear damping of nuclear spin waves in antiferromagnetic $CsMnF_3$

A.V. Andrienko

I. V. Kurchatov Institute of Atomic Energy (Submitted 18 November 1991) Zh. Eksp. Teor. Fiz. 101, 1644–1662 (May 1992)

The system of parametric nuclear spin waves (NSWs) in the region of "hard" excitation was investigated in detail. The effect of the modulation of the magnon spectrum at the thresholds for appearance (h_{c1}) and quenching (h_{c2}) of parametric magnons were studied under conditions of parallel pumping. It was found that the peak at the threshold h_{c1} with pump frequency $v_p \approx 780$ MHz is due to the resonance absorption of magnons by defects having a transition frequency $v_{def} \approx 390$ MHz, while hard excitation of NSWs is governed by the turning off of the corresponding relaxation mechanism when transitions with frequency $v_p/2$ are saturated. The relaxation time of the defects responsible for the hardness was measured under conditions of weak and strong excitation of nuclear magnons. Simultaneous measurements of the antiferromagnetic and nuclear magnetic resonance spectra were performed under conditions of excitation of NSWs. It was determined that in the case when parametric excitation of NSWs decreases the nuclear magnetization and the nuclear subsystem is in a state of flux equilibrium, the spectra of the coupled electronic-nuclear oscillations are not described by the theory of de Gennes *et al.* [Phys. Rev. **129**, 1105 (1963)].

In weakly anisotropic antiferromagnets a new branch of coupled oscillations of electronic and nuclear spins—nuclear spin waves (NSWs)—arises as a result of the hyperfine interaction between the nuclear spins and electronic shells of the magnetic ions.¹ The frequencies of these oscillations lie in the NMR frequency range ($\nu_n \leq 700$ MHz for ⁵⁵Mn nuclei) and at wave-vectors $\mathbf{k} \leq 10^6$ cm⁻¹.

Inhomogeneous oscillations of this branch of NSWs or magnons can be excited parametrically by applying to the sample a microwave magnetic field $h \cos \omega_p t$ parallel to a static magnetic field H. When the amplitude of the microwave field exceeds some critical value h_c , parametric instability with respect to the decay of a microwave pump photon into two magnons with frequencies $\omega_k = \omega_p/2$ and with equal and oppositely directed wave vectors k and $-\mathbf{k}$ develops in the sample. The critical amplitude of the microwave field is related linearly with the relaxation Γ of the excited NSWs (magnons), $h_c V = \Gamma$, where V is the coupling constant between the magnons and the pump field.²

Investigation of parametric excitation of nuclear magnons in CsMnF₃ showed³ that the excitation in the region of pump frequencies $\omega_p/2\pi = v_p = 700-900$ MHz is hard: There exist two threshold fields h_{c1} and h_{c2} ($h_{c1} > h_{c2}$), corresponding to appearance and quenching, respectively, of parametric magnons. The frequency dependence of the hardness parameter

$$\xi = h_{c1}/h_{c2} - 1$$

was investigated in Ref. 4. It was found that $\xi(v_p)$ has a pronounced resonance character, and to within 2 MHz the position of the peak corresponding to $v_p = 782 \pm 2$ MHz is independent of temperature and does not vary from sample to sample, though the quantity ξ does vary. The fact that ξ varies from sample to sample indicates that the observed phenomenon is caused by defects in the crystals.

The hardness can be explained by the following qualitative arguments. Suppose that some defect in the specimen consists of a two- or multilevel system with characteristic transition energy $\varepsilon = \hbar \omega_p / 2$ (or $\hbar \omega_p$). Such a system can absorb magnons (pairs of magnons) resonantly and in the process pass into an excited state. However, when the number of parametric NSWs is large the system becomes saturated (the difference of the populations of the levels is equal to zero) and the magnon damping decreases. Such "negative nonlinear damping" could become responsible for the hard character of the excitation of NSWs. Then the physical parameter characterizing the hardness will be not ξ but rather the turning-off part of the relaxation $\Delta \Gamma = (h_{c1} - h_{c2}) V$.

Although at the present time this is virtually the only qualitative model that can explain the hardness of NSWs, it has still not been confirmed experimentally. For this reason, the aim of the present work was to investigate in detail parametric excitation and relaxation of nuclear magnons in the region of hard excitation.

EXPERIMENTAL PROCEDURE

Parametric NSWs were excited by parallel microwave pumping with frequency $\omega_p/2\pi = 700-900$ MHz. A CsMnF₃ single crystal with a volume of 10-20 mm³ was placed at the center of a spiral copper resonator 9 mm in diameter (Fig. 1), which was essentially a half-wave dipole coiled into a spiral. An antenna was used for input coupling with the resonator. At critical input coupling the loaded Q at liquid-helium temperatures was equal to $Q_L \sim 500$. The microwave magnetic field of the resonator was directed along the axis of the spiral. A modulation coil 20 mm in diameter was wound coaxially with the resonator, and a signal, which sinusoidally modulated the static magnetic field at a frequency $\omega_m/2\pi = 1-700$ kHz, was fed into the modulation coil. Weak modulation of the magnetic field was employed to record the parametric excitation, and strong modulation was employed to investigate the effect of modulation on the pumping threshold and on the supercritical behavior of the magnons. In most experiments the static field was directed

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FIG. 1. Schematic diagram of the absorbing cell and the direction of the magnetic fields relative to the C_6 axis of the crystal: 1) Spiral resonator, 2) loop, 3) modulation coil, 4) sample, 5, 6, 7) coaxial waveguides from the pump generator, the receiving channel, and the probe-pump generator, respectively.

along the resonator axis, i.e., the condition $H \mid h \mid H_m$ was satisfied.

A probe pump was applied to the sample in addition to the main pump. For this, a loop, which shorted the coaxial cable connected with the probe-pump generator, was inserted at the center of the resonator. The plane of the loop was parallel and the microwave magnetic field generated at the sample by the loop was perpendicular to the resonator axis and the main pump field $(h_3 \perp h \parallel H)$. At this geometry of the experiment the loop has virtually no effect on the Q of the resonator, and this made it possible to study the response of the sample to two microwave fields. The probe-pump frequency was varied in the range $\omega_{\rm pr}/2\pi = 200-9000$ MHz. The signals from all microwave generators were detected with an antenna connected to a receiver and a crystal detector.

In individual cases it was necessary to increase the amplitude of the probe pump and to increase the sensitivity of the receiving circuit to the signal generated by probe generator and absorbed by the sample. Then the coaxial line, shorted by the loop, was disconnected from the generator and they were coupled with the help of a capacitor soldered to the central conductor. The coaxial resonator created in this manner with the loop at the end had a natural frequency of the order of 150 MHz, so that the higher-order harmonics of the resonator were employed. The frequency of the resonator could be tuned over wide limits with the help of a variable-length line placed in the resonator. At frequencies of the order of 800 MHz the Q of this resonator was $Q_L \sim 100$.

The modulation response of the sample was used to record the parametric excitation of the nuclear magnons. As shown in Refs. 3 and 5, when the energy spectrum of spin waves is modulated by an oscillating magnetic field $H_m \cos(\omega_m t)$ the microwave pump signal which passes through the resonator is modulated at the frequency ω_m . The modulation response and hence also the parametric excitation of the magnons were recorded with a spectrum analyzer placed at the output of the microwave receiver.

The values of the microwave field h in the spiral resonator were calculated from the microwave power fed into the resonator, the Q of the resonator, and the input coupling.³ The value of h_c was determined with an absolute error of 25%; the relative error did not exceed 2%.

In the experimental investigation of the effect of parametric excitation of NSWs on the antiferromagnetic resonance (AFMR) spectrum the signal of the probe pump at the frequency $\omega_{\rm pr}/2\pi = 6-9$ GHz was detected with a crystal detector and recorded on an X-Y plotter as a function of the dc magnetic field.

The CsMnF₃ sample was oriented in the measuring cell in a manner (Fig. 1) so that the principal axis C_6 of the sample was oriented vertically and all oscillating magnetic fields and the static magnetic field lay in the basal plane of the crystal, which is also the easy-magnetization plane.

EFFECT OF MODULATION ON THE PUMPING THRESHOLDS UNDER CONDITIONS OF HARD EXCITATION OF NSWs

In the first series of experiments we studied the effect of the modulation of the NSW spectrum on the thresholds h_{c1} and h_{c2} . According to the theory,^{6,7} the effect of the modulating field $H_m \cos \omega_m t$ on the parallel-pumping threshold under conditions such that $\omega_p/2\pi \gg \omega_m/2\pi > 2\Gamma$ and $h_c/h_{c0} - 1 \ll 1$ is described by the expression

$$\frac{h_e}{h_{eo}} - 1 = 4V^2 H_m^2 [v_m^2 + 4\Gamma^2]^{-1}.$$
 (1)

Here h_{c0} and h_c are the threshold fields in the absence and presence of modulation, respectively.

If the hardness is explained with the help of the model of the "switchable part of the relaxation," then it should be expected that the effect of modulation of the threshold fields h_{c1} and h_{c2} will be different because the magnon relaxation rate is different at the moment these thresholds are measured:

$$\Gamma_1 = h_{c1}V > \Gamma_2 = h_{c2}V$$

It is obvious that the effect on the threshold h_{c1} should be much smaller than the effect on the threshold h_{c2} . In addition, the effect of modulation on the threshold is a maximum when $v_m \sim 2\Gamma$,⁷ i.e., the maximum of $(h_c/h_{c0} - 1)$ as a function of v_m makes it possible to estimate the relaxation rate of the excited waves which corresponds to both pumping thresholds.

The experiments showed that the effect of H_m on the thresholds h_{c1} and h_{c2} is significantly different and that this difference increases with increasing ratio h_{c1}/h_{c2} , which can be easily controlled by varying the temperature or pump frequency. The fact that Γ_1 is different from Γ_2 has the effect that for modulation amplitudes such that the relative increase in the threshold h_{c1} is still small, the strength h_{c2} of the main pump increases so much that combinational pumpings of nuclear magnons at the frequencies $\omega_p \pm \omega_m$, $\pm 2\omega_m$, and so on, whose thresholds are found to be lower than the threshold of the main pump, start in the samples. This effect is manifested in the fact that maxima corresponding to the limits of existence of different pumps appear in the ratio h_{c2}/h_{c20} as a function of the maximum frequency v_m (Ref. 7).

Figure 2 shows the experimental results for the relative increase in the thresholds h_{c1} and h_{c2} under conditions of comparatively low hardness, i.e., for $h_{c10}/h_{c20} = 2.16$. In the hardness model considered here this means that the ratio of the relaxation parameters is the same. Figure 2 confirms the model of turning-off part of the relaxation, since both the relative increase of the pumping thresholds and the positions of the principal maxima in the frequency dependences of the



FIG. 2. Effect of modulation on the pumping threshold in the region of low hardness: $h_{c10}/h_{c20} = 2.16$; $v_p = 779.37$ MHz, T = 4.2 K, H = 0.725 kOe, $H_m = 2.8$ Oe; $\bigcirc -h_{c2}$, $\blacksquare -h_{c1}$.

thresholds agree satisfactorily with the values of Γ_1 and Γ_2 calculated from the thresholds h_{c10} and h_{c20} : $\Gamma_1 = 173 \pm 43$ kHz and $\Gamma_2 = 80 \pm 20$ kHz.

The study of the effect of the modulation of the energy spectrum of NSWs under conditions of high hardness gives an even more striking picture. Figure 3 shows these results in the case $h_{c10}/h_{c20} = 5.65$. The colossal difference in the magnitude of the effect of modulation on the thresholds h_{c1} and h_{c2} and in the position of the maxima also correspond to the relaxation rates calculated from the two threshold fields.

Thus the existence of two threshold fields is indeed due to the decrease in the relaxation rate of parametrically excited nuclear magnons. This completely confirms the negative nonlinear damping hypothesis.

We also note the following detail. For high modulatingfield frequencies $v_m \ge 500$ kHz a high modulation amplitude H_m gives rise not to an increase but rather a decrease of the parametric-excitation threshold h_{c1} . This evidently happens because the relaxation rate of magnons with frequency $(\omega_p \pm \omega_m)/2$ is significantly (by more than a factor 2) lower than the relaxation rate of magnons with frequency $\omega_p/2$, which correspond to maximum hardness. As a result, combinational pumping starts at a lower amplitude of the microwave magnetic field than the main pumping. Therefore in this case we measure the effect of modulation not on h_{c10} but rather on the threshold of combinational pumping. This phenomenon has not been observed before, since it can arise



FIG. 3. Effect of modulation on the pumping thresholds in the region of high hardness: $h_{c10}/h_{c20} = 5.65$; $v_p = 778.7$ MHz, T = 1.76 K, H = 1.08 kOe; $H_m = 1.96$ Oe; $\bigcirc -h_{c2}$, $\frown -h_{c1}$.

only if the maximum in the relaxation of the excited waves is very sharp.

Aside from the effect of the field H_m on the pumping thresholds, we also observed a number of new effects associated with the effect of modulation on the supercritical behavior of parametric magnons.

It is well known that when the modulating-field amplitude $H_m \cos(\omega_m t)$ is large, a response at the frequency $\omega_m/2$ appears together with a modulation response at the frequency ω_m at a given threshold. This effect was first observed in yttrium-iron garnet⁶ and has been termed double parametric resonance. In Ref. 7 it was shown that in the system of parametric nuclear magnons in CsMnF₃ there exist several regions in the (h, H_m) plane in which signals are observed simultaneously at ω_m and $\omega_m/2$ (the measurements were performed at $\omega_m = 2\pi \cdot 80$ kHz. As H_m is increased a transition to chaotic behavior can theoretically occur through successive period doubling of the oscillations.⁸ In Ref. 7, however, the transition to chaos occurred above the threshold of double parametric resonance without the appearance of subharmonics at the frequencies $\omega_m/4$, $\omega_m/8$, etc.; this is apparently attributable to the effect of fluctuations.

In the present work we were able to observe in the spectrum of a microwave signal which has passed through the resonator signals with frequencies $\omega_m/2$, $\omega_m/4$, and $\omega_m/8$ and even a modulational response with frequency $\omega_m/3$. The fundamental difference from Ref. 7 lies is that these signals were observed only at low modulation frequencies $v_m < 3$ kHz (the largest number of subharmonics appearing simultaneously was observed at the frequency $v_m = v^* = 2$ kHz), i.e., when the modulation period is much shorter than the lifetime of parametric waves $\tau_m \sim 10^{-5}$ s (T = 1.76 K) and the inverse frequencies of the collective oscillations. Thus, in contrast to Ref. 7, where a dynamic regime of transition to chaos (Feigenbaum's scenario) was observed, in the present case chaos apparently develops in a manner similar to hydrodynamic turbulence.

Another interesting effect associated with the anomalous behavior of the system of NSWs at the frequency v^* was



FIG. 4. Spectrum of the modulation response of the system of NSWs near the frequency $\omega_m/2$ in the presence of two modulating fields with close frequencies: $\omega_{m1} = 2\pi \cdot 60$ kHz and $\omega_{m2} = 2\pi \cdot 64$ kHz. The spectra 1-4 correspond to different modulation amplitudes; the modulation increases from the first to the fourth spectrum. T = 1.82 K, H = 1 kOe, $\omega_e = 2\pi \cdot 779.2$ MHz, $H_{m1} \approx H_{m2}$.

observed when two modulating signals at frequencies of the order of the frequency of the collective oscillations $v_{m1} = 60$ kHz and $v_{m2} = 64$ kHz, such that $(v_{m2} - v_{m1})/2 \approx v^*$, were applied to the sample. As the amplitudes H_{m1} and H_{m2} were increased, signals at $v_{m1}/2$ and $v_{m2}/2$ appeared first at a certain threshold (curve 1 in Fig. 4). As the amplitudes were increased further, a signal with frequency (v_{m1}) $(v_{m2})/2 = 31$ kHz was added (curve 2). As H_m increased this signal increased and the double-resonance signals vanished at a certain threshold (curve 3). When H_m was raised above the next threshold value, the modulational response with frequency $(v_{m1} + v_{m2})/2$ vanished, but near this frequency there appeared an entire series of signals, separated by $\frac{1}{3}$ kHz from the frequencies $v_{m1}/2$ and $v_{m2}/2$. We note that all these effects are associated with the hardness, since they exist only in the region of hard excitation of NSWs.

INVESTIGATION OF THE HARDNESS PARAMETERS

Since investigations of the effect of modulation on the pumping thresholds confirmed the proposition that the hardness of parametric excitation of nuclear magnons is caused by negative nonlinear damping, we focus in the hardness study below on measurements of the shutoff part of the relaxation of nuclear magnons.

We investigated in detail the frequency, field, and temperature dependences of the negative nonlinear damping. The frequency dependence of $\Delta\Gamma$ was measured in a tunable coaxial resonator. Figure 5 shows the frequency dependence of the shutoff part of the nuclear-magnon relaxation rate $\Delta\Gamma$ in one of the CsMnF₃ samples at a fixed magnetic field *H*. A similar dependence constructed for a fixed magnon wave vector **k** is virtually identical to that shown in Fig. 5. On the basis of the model under study, the width of the peak of the temperature dependence $\Delta\Gamma(T)$ can correspond either to the defect relaxation rate or to the frequency distribution of defects near $v_{def} \approx 391$ MHz. Since the width of the peak in the hardness ξ is virtually temperature independent,⁴ in the first case this would mean that the lifetime of the excited



FIG. 5. Frequency dependence of the turning-off part of the relaxation in $CsMnF_3$. T = 1.86 K, H = 0.8 kOe.



FIG. 6. a) Hardness parameter $\xi = h_{c1}/h_{c2} - 1$ as a function of the temperature at the peak of the frequency dependence of $\Delta\Gamma$ at H = 670 Oe; b) temperature dependence of the turning-off part of the relaxation $\Delta\Gamma$; H = 670 Oe, $\nu_p = 780$ MHz.

defect is independent of T, which is unlikely.

Figure 6 shows for comparison the temperature dependences of both the parameter ξ and the shutoff part of the relaxation $\Delta\Gamma$ at maximum hardness. It was found that as the temperature increases ξ decreases while the shutoff part of the relaxation increases, and $\Delta\Gamma \propto T$ in a significant fraction of the temperature range studied. The quantity $\Delta\Gamma$ passes through a maximum at $T \approx 3.5$ K. The possible cause of the incipient decay of $\Delta\Gamma$ at high temperatures could be that the difference in the defect-level population decreases with increasing temperature and that these levels no longer contribute to the magnon damping.

The quantity $\Delta\Gamma$ depends quite weakly on the magnetic field H (Fig.7a). If these results are replotted in coordinates $\Delta\Gamma(k)$, we find that the shutoff part of the relaxation increases with increasing magnon wave vector (Fig. 7b), as is characteristic for interaction of NSWs with defects.^{4,9}



FIG. 7. a) Turning-off part of the relaxation $\Delta\Gamma$ versus the magnetic field. T = 1.93 K, $\nu_p = 784$ MHz; b) same dependence in $\Delta\Gamma$ (k) coordinates. k is in the 10^5 cm⁻¹ units.

HARDNESS SHUTOFF BY SATURATION OF DEFECTS BY MICROWAVE PROBE RADIATION

To obtain additional information on the nature of the hardness and on the phenomena occurring in the magnetic system, we performed a series of measurements in which, aside from the main pumping with frequency $v_p \approx 780$ MHz, a microwave probe was applied to the sample.

In the first run of experiments the frequency $v_{\rm pr}$ of the probe pump was of the order of v_p . The microwave magnetic field of this pump was perpendicular to the constant field $(h_{\rm pr} \perp H)$. When the condition $v_{\rm pr} = v_p/2$ was satisfied, the second pump significantly affected the threshold h_{c1} of the main pump. This effect started at amplitudes $h_{\rm pr} \sim 3 \cdot 10^{-4} h_{c1}$. As the probe-pump power $P_{\rm pr}$ increased the threshold h_{c1} decreased, approaching the value h_{c2} , i.e., the shutoff part of the relaxation $\Delta\Gamma$ approached zero (Fig. 8). No effect of the probe with frequency $v_{\rm pr} = v_p/2$ on the threshold field h_{c2} was observed.

The decrease of $\Delta\Gamma$ can be explained on the basis of the proposed model by assuming that the populations of the levels of the defects with activation energy $\varepsilon = \hbar \omega_{\rm pr} = \hbar \omega_p / 2$ become equalized. The $\Delta\Gamma(P_{\rm pr})$ dependence is not continuous. Two strong jumps of $P_{\rm pr}$ are observed at T = 1.5 K. At the jumps, the power $P_{\rm pr}$ increases abruptly by a factor 10–40 for some values $\Delta\Gamma = \Delta\Gamma_b$. For example, very-low-power microwave radiation saturating the levels of the defects is sufficient to reduce the negative nonlinear damping of NSWs from $\Delta\Gamma$ to some value $\Delta\Gamma_b \approx 0.2\Delta\Gamma$, but to reduce $\Delta\Gamma$ below $\Delta\Gamma_b$ the probe power $P_{\rm pr}$ must be increased abruptly by a factor of 40 ($P_{\rm pr} \ll P_{\rm c1}$ as before). This circumstance suggests that the system of defects responsible for $\Delta\Gamma$ has a quite complicated structure, including several charac-



FIG. 8. Turning-off part of the relaxation as a function of the probe pump power with frequency $v_{\rm pr} = v_{\rm p}/2 = 391.00$ MHz; O—T = 4.2 K, H = 750 Oe; Φ —T = 1.5 K, H = 1120 Oe. The arrows mark the values of $\Delta\Gamma$ for $P_{\rm pr} = 0$.



FIG. 9. Probe-pump power necessary for exciting magnons with frequency $v_k = v_p/2 = 390.35$ MHz as a function of the probe-pump frequency for different main-pump amplitudes $h: h/h_{cl} = 0.4(\Delta), 0.44(\Phi), 0.88(\bigcirc)$. T = 1.5 K, H = 1120 Oe.

teristic relaxation times. We note that the field amplitude $h_b = (\Gamma_2 + \Delta \Gamma_b)/V$ is the threshold below which the stationary state of parametric magnons becomes unstable, i.e., low-frequency self-excited oscillations exist, and in addition the threshold h_b is not related with the presence of the probe pump field.

When the frequency $v_{\rm pr}$ is shifted from $v_p/2$ the effect on the threshold decreases rapidly. The experiment was performed as follows. The power $P_{\rm pr}$ at fixed v_p , $v_{\rm pr}$, and $h(h_{c2} - h - h_{c1})$ was increased continuously until pumping started. The value of $P_{\rm pr}$ was measured. Next, the frequency $v_{\rm pr}$ was changed and the power $P_{\rm pr}$ corresponding to it measured. Thus $P_{\rm pr}$ in Fig. 9 is the power required to lower the threshold h_{c1} to the value h, i.e., the power required to turn off some fixed part

$$\Delta\Gamma(h_{c1}-h)/(h_{c1}-h_{c2})$$

of the relaxation of the nuclear spins. The resonant character of the effect of $P_{\rm pr}$ on $h_{\rm cl}$ can be seen clearly. Just as in Fig. 8, a strong jump in the power (by two orders of magnitude) is observed when $h/h_{\rm cl}$ is changed from 0.40 to 0.44.

If it is assumed that the linewidth at the level $2P_{\rm pr,min}$ characterizes the characteristic relaxation rate of the defect, then we obtain the values $\Delta v(T = 4.2 \text{ K}) \approx 20 \text{ kHz}$ and $\Delta v(T = 1.5 \text{ K}) \approx 10 \text{ kHz}$. We note that these values are significantly (by two orders of magnitude) smaller than the width $\Delta \Gamma$ of the peak in Fig. 5; the latter width is probably determined by the frequency distribution of the defects.

In Fig. 9 it is obvious that the line $P_{\rm pr}(v_{\rm pr})$ exhibits some asymmetry and its width increases with increasing *h*. The observed broadening is apparently explained by the fact that defects having a transition frequency $v_{\rm pr}$ absorb some of the power $P_{\rm pr}$ and then emit magnons whose frequency changes slightly after a series of quasielastic¹ interactions with fluctuations of the nuclear magnetization^{10,11} (they "diffuse" over the NSW spectrum) and which are once again absorbed by defects, but now with different frequencies, including also $v_p/2$. This results in saturation of the defects not by the probe pump, but rather by the secondary magnons. Since one defect can emit T_1^{-1} magnons per second (T_1 is the lifetime of the excitation of a defect), the number of such secondary magnons is limited, so that their contribution to the decrease in $\Delta\Gamma$ is most noticeable for $h \sim h_{c1}$, i.e., when the subsystem of defects with frequency $v_p/2$ need be saturated only partially to realize parametric excitation of the magnons.

The effect of probe radiation with frequency $v_{\rm pr} = v_p$ on the pumping thresholds was also investigated. It was observed that this radiation has a small effect on both thresholds, but the effect becomes appreciable only when the probe-pump amplitude $h_{\rm pr}$ is equal to the amplitude of the main pump. It is natural to conjecture that the effect of $P_{\rm pr}$ on the thresholds h_{c1} and h_{c2} is governed by the presence of the projection of the field $h_{\rm pr}$ on the direction of the static field H, i.e., the effect of $P_{\rm pr}$ on the thresholds is a consequence of the fact that the main pump power is increased with the help of an additional microwave generator and not by saturation of defects by the probe pump.

We note that the effect of the additional irradiation on the hardness of the parametric excitation was investigated previously for electronic spin waves in CsMnF₃ (Ref. 12) and in the weak ferromagnet FeBO₃ (Ref. 13). In addition, it was observed in Ref. 12 that parametric magnons with frequency $\omega_{p2}/2$ affect the hardness of a pump with frequency ω_{p1} if the frequencies of the excited magnons do not differ by more than 1.5 GHz. This result is superficially reminiscent of the result obtained in the present work, but the hardness is shut off not because of direct saturation of defects by the additional pump field but rather as a result of an increase in the number of magnons near the frequency $\omega_{p1}/2$, while for the electronic spin waves the hardness parameter itself is not resonant.

INVESTIGATION OF THE RELAXATION TIME OF THE SYSTEM OF DEFECTS

Figure 10 shows schematically the number of parametric magnons as a function of the pump amplitude in the region of hard excitation. It is clear that in the hardness range $h_{c2} < h < h_{c1}$ the system of parametric NSWs can be in one of two states: 1) unexcited or 2) excited. The state of the magnon system depends on its previous history. This makes pos-



FIG. 10. Number of parametric magnons as a function of the microwave pump amplitude under conditions of hard excitation (schematically).

sible a study of the transitions of the system of magnons from state 2 into state 1.

The experiment was organized as follows. The microwave magnetic field in the continuous generation mode was raised above h_{c1} to generate parametric magnons. The field was then lowered below h_{c1} to some value h at which an excited state 2 was established in the system (point C in Fig. 10). Next, a blocking pulse of width τ^* was fed to a *pin*-diode and blocked the microwave channel. The attenuation of the microwave power flowing into the resonator with the sample in the closed channel was at least 30 dB. The microwave channel was then opened and the receiving system recorded whether the state 2 was restored after the microwave pump was turned on again or the magnon system was transferred into the state 1. We investigated the minimum pump interruption time τ for which parametric excitation of magnons was not restored and the magnon system was transferred from state 2 into state 1. The measurements were performed at two pump frequencies:

 $v_p = 784$ MHz, i.e., at peak hardness,

and

 $v_p = 764$ MHz, i.e., for $\xi = 0.1 \xi_{\text{max}}$.

The results were similar. The characteristic $\tau(h)$ dependences are presented in Fig. 11. As one can see from this figure, for h in the interval $h_{c2} < h < h_{c1}$ the quantity τ changed by several orders of magnitude and reached 10 s in a number of experiments. Thus the magnetic system of the sample remembers the parametric nuclear magnons for a time much longer than the lifetime of the excited quasiparticles ($\tau_m \approx 1 \, \mu$ s at T = 4.2 K and $\tau_m \approx 10 \, \mu$ s at T = 1.65 K). In addition, the maximum times τ correspond approximately to the spin-lattice relaxation times of the average magnetization of Mn nuclei.¹⁴ In Fig. 11 one can see the following interesting feature: For some value of h the time τ increases abruptly by a factor of $10^2 - 10^3$, i.e., it has the same nonmonotonic character as the power $P_{\rm pr}$ required to saturate the defects.



FIG. 11. Minimum pumping interruption time τ required in order to transfer the system of parametric magnons from the excited into the unexcited state, as a function of the microwave field amplitude *h*. The arrows mark the positions of the threshold fields: $\uparrow -h_{c2}$, $\downarrow -h_{c1}$. $\nu_p = 784$ MHz. $\Theta - T = 1.65$ K, H = 770 Oe; $\triangle - T = 4.2$ K, H = 770 Oe; $\bigcirc - T = 4.2$ K, H = 760 Oe.

We now analyze the experimental results in the model of negative nonlinear damping of NSWs for the case of a multilevel system of defects. Let $\Gamma_2 = h_{c2} V$ be the characteristic magnon relaxation rate and let $\Delta \Gamma = \Gamma_{def} \langle S^z \rangle^n$ be the contribution of defects to magnon relaxation (we employ in the calculation a spin model with n + 1 projections on some axis Z). When the defects are overheated by the parametric NSWs the temperature of the system approaches infinity $T_{def} \rightarrow \infty$ (all levels have the same population), and the contribution to magnon relaxation $\Delta\Gamma(T_{def}) \rightarrow 0$. In addition, $h_{c1} \rightarrow h_{c2}$. When the pumping is interrupted the system of defects cools according to the law $\langle S^z \rangle = \langle S^z \rangle_0 [1 - \exp(-t/T_1)]$. At the same time, $\Delta \Gamma$ increases. If the temperature of the defects drops in the time τ to such an extent that $\Gamma_2 + \Delta \Gamma$ $(\tau, T_1) > hV$, the field h is then unable to excite the pump, and the magnon system passes into the state 1. On the basis of the proposed model, we obtain the following expression relating the parameters of the experiment with the relaxation time T_1 of a defect:

$$\frac{\tau}{T_{1}} = -\ln\left\{1 - \left[\frac{h - h_{c2}}{h_{c1} - h_{c2}}\right]^{1/n}\right\},$$
(2)

where n = x - 1 for an x-level system. The experimental results for $\tau \leq 50 \,\mu s$ (before the jump in the value of τ) are best described by Eq. (2) with n = 2 (Fig. 12), i.e., a three-level system with relaxation time $T_1 \approx 10 \ \mu s$ at $T = 4.2 \ K$ and $T_1 \approx 30 \,\mu s$ at T = 1.65 K. In addition, within the limits of the experimental error the relaxation rate T_{1}^{-1} of the three-level system does not depend on the magnetic field and is proportional to the temperature of the sample. If the lifetime T_1 is converted into the relaxation rate of the system, we obtain $\Delta v_{def} = 1/2\pi T_1 \approx 16$ kHz at T = 4.2 K and $\Delta v_{def} \approx 6$ kHz at T = 1.65 K, which correspond fairly well to the linewidth Δv obtained for the defects from experiments on saturation of the defects by the probe pump. We note that the contribution of the defects to the magnon relaxation exhibits the same temperature dependence $(\Delta \Gamma \propto T)$ as the relaxation rate Δv_{def} of the system of defects itself.

The theory does not describe the strong increase in τ as *h* increases further. A similar effect—slow increase in the threshold h_{c1} of parametric excitation of electronic magnons in FeBO₃ after the saturating pulse is turned off—was also observed in Ref. 13, where the characteristic time τ was 30 times longer than the lifetime of the parametric magnons. In



FIG. 12. Data from Fig. 11 in the $\tau(X)$ coordinates, where $X = -\ln\{1 - (h - h_{c2})^{1/2}/(h_{c1} - h_{c2})^{1/2}\}$. $\tau = T_1$ for X = 1.

Ref. 13 it was suggested that the saturating pulse excites characteristic elastic vibrations of the sample, which have a lifetime of up to 500 μ s and affect the hardness of the parametric excitation.

In our case, the observed times τ are five to six orders of magnitude longer than the magnon relaxation times. The reason for this may be the existence of several defect relaxation times or the fact that the time T_1 depends on the extent to which the nuclear system as a whole or near the frequency $\omega_p/2$ is overheated. It is obvious that if the temperature of the Mn nuclei is high, the cooling rate of the defects will be determined by the spin-lattice relaxation time $T_{\rm sl}$ of the nuclear system, which is the longest time in the magnet studied. According to Ref. 14, $T_{\rm sl} \approx 0.1$ s at T = 4.2 K and $T_{\rm sl} \approx 3$ s at T = 1.65K, which agrees well with the maximum values of τ in experiments on interruption of pumping.

We undertook another series of experiments designed to determine the relaxation time of the defects. The basic idea of these experiments was to perform measurements in the absence of overheating of the nuclear system by parametric magnons; this overheating could affect T_1 . In the first series of experiments the sample was irradiated with the pump ω_{p1} in the continuous generation regime with amplitude $h_{c2} < h < h_{c1}$ and with narrow (less than 1 ms in width) powerful probe-pump pulses with frequency $\omega_{pr} = \omega_{p1}/2$. The pulse width was chosen so that a single pulse could not overheat the system of defects to such an extent that parametric excitation would start, though the periodic action of the probe pulses ω_{pr} resulted in parametric excitation of NSWs even when the repetition frequency was equal to 5 Hz. This means that there is not enough time for defects to relax between the pulses, i.e., the defect relaxation time in the absence of parametric NSWs at temperature T = 4.2 K corresponds completely to the maximum time $\tau \sim 0.1$ s obtained in experiments on interruption of pumping in which the nuclear system was strongly excited. Further investigations showed, however, that in these experiments the field of the main microwave pump affected the relaxation time of the defects.

In subsequent experiments with the same temperature T and magnetic field H the defect cooling time was measured not only in the absence of parametric excitation, but also with the main pump field ω_{p1} turned off. First, the sample was irradiated only with a powerful probe-pump pulse, sufficient for saturating the system of defects, i.e., for lowering the pump threshold h_{c1} to h_{c2} . Then the probe-pump pulse was followed after a time interval t by the main-pump pulse with amplitude $h_{c2} < h < h_{c1}$, which excited parametric magnons if t was sufficiently short. By increasing the time interval t between these pulses we found that over a time $t \sim 200$ μ s the pumping threshold reaches the value h_{c1} , i.e., there was enough time for the defects to relax to the lattice temperature. Thus it was established experimentally that the relaxation rate of the defects decreases not only in the presence of parametric magnons, but also in the presence of a microwave magnetic field of frequency $\omega_p = 2\omega_{def}$.

INVESTIGATION OF ANTIFERROMAGNETIC AND NUCLEAR MAGNETIC RESONANCE SPECTRA IN THE PRESENCE OF PARAMETRIC NSWS

As shown in the preceding section, the relaxation time of defects far above threshold approximately corresponds to the spin-lattice relaxation time of the nuclei. This suggests that the entire nuclear system is significantly overheated. For this reason, the aim of the next series of experiments was to measure the temperature of the nuclear magnetic subsystem under conditions of parametric excitation of NSWs.

It is well known that when the nuclear temperature changes the spectra of nuclear and electronic spin waves (ESWs) also change. These changes are most easily determined from the recorded shift of the antiferromagnetic resonance (AFMR) and nuclear magnetic resonance frequencies. The calculation of the spectra of coupled electronic-nuclear oscillations, which was presented in Ref. 1, gives the following expressions for CsMnF₃:

$$\omega_{c}^{2} = \gamma^{2} (H^{2} + H_{\Delta}^{2}),$$

$$\omega_{n}^{2} = \omega_{n0} (1 - \gamma^{2} H_{\Delta}^{2} / \omega_{c}^{2}).$$
(3)

Here ω_e is the AFMR frequency, ω_{n0} is the unshifted NMR frequency in the hyperfine field of the electron shell, γ is the magnetomechanical ratio, γH_{Δ} is the hyperfine gap in the AFMR spectrum, and $H^2_{\Delta} = (6.4/T)$ kOe² is proportional to the average nuclear magnetization $\langle m \rangle$, i.e., according to Curie's law, it is inversely proportional to the temperature of the nuclear system.

In the first experiment we studied the dependence of the AFMR field on the power pumping the nuclear magnons at fixed frequency. The signal passing through the resonator was recorded with the magnetic field scanned from H = 0 up to H = 1.6 kOe. The scan rate was equal to ~200 Oe/s. It was found that the shift of the AFMR line becomes noticeable even with supercriticality $h/h_{c2} \approx 2$, i.e., approximately for the same values of h at which a jump occurs in the time τ in Fig. 11. As the supercriticality increases, the resonance field H_e increases up to the values of the limiting pump field H_b (Fig. 13, curves l and 2). To estimate the rate of cooling of the nuclear system we set the field at $H \approx H_b$ and then abruptly turned off the nuclear pump and the static magnetic field (over a time ~0.1 s), and after some time we scanned



FIG. 13. Curves *l* and 2 represent the dependence of the position of the AFMR line with frequency $v_e = 6189$ MHz on the amplitude of microwave pumping of nuclear magnons with frequency $v_k = v_{p1}/2 = 391$ MHz at T = 1.55 K. 1) $P(v_p) = 0$; $2)P(v_p) \approx 0.1$ W. The curves 4 and 5 represent the limits of the microwave pumping of NSWs with frequency $v_{k2} = v_{p2}/2 = 375$ MHz under the same conditions as curves *l* and 2, respectively. Curve 3 is the AFMR line 10 s after the parametric NSWs have cooled in a field H = 0.

the field at a rate ~ 500 Oe/s. We found that the AFMR line returns from the field $H \approx H_b$ to the initial position within a time ~ 100 s. Figure 13 shows the AFMR line after the nuclear system cools in a zero field over a time 10 s (curve 3).

According to the formula (3), the observed shift of the AFMR line (curve 2) corresponds to a decrease of the average nuclear magnetization by a factor of 1.5, i.e., increase of the nuclear temperature to 2.26 K. In the present experiment a change in the nuclear magnetization is recorded only in the field H_e , though it is possible that for other values of the field it is even larger. Under conditions of thermodynamic equilibrium, such an increase of the temperature of the nuclei would result in an appreciable change of the NMR spectrum. Calculation using (3) shows that at the temperature T = 2.26 K the limiting pump field of frequency $v_p = 782$ MHz is equal to $H_b = 1.22$ kOe, so that in a magnetic field $H_e = 1.54$ kOe the frequency $\omega_p/2 = 2\pi \cdot 391$ MHz should lie significantly below the bottom of the NSW band, i.e., theoretically, AFMR with frequency 6129 MHz and nuclear magnons with frequency 391 MHz cannot exist simultaneously in the field H = 1.45 kOe. This means that under the conditions of strong excitation of NSWs the theory of Ref. 1 does not describe the spectrum of coupled electronic-nuclear oscillations.

A similar shift of the AFMR line was observed in Ref. 15 under conditions of parametric excitation of NSWs in antiferromagnetic MnCO₃. The pumping limit remained the same. The authors of Ref. 15 paid no attention to this latter circumstance; attributing the shift of the AFMR line to overheating of the nuclear system, they ignored the consequent contradiction. Our estimates, based on the results of Ref. 15, show that the observed shift of the AFMR line from the field H = 1.16 kOe to the field H = 1.54 kOe corresponds to an increase in the temperature from 1.4 to 4.03 K. With this overheating the limit of pumping of NSWs should be in the field $H_b = 1.05$ kOe, i.e., just as in our case, it is impossible to observe simultaneously AFMR and pumping of NSWs in a field H = 1.54 kOe. Therefore the theoretical calculations performed in Ref. 1 cannot be used to describe these experimental results.

To estimate how much the NMR frequency is changed by parametric excitation of nuclear magnons, we performed an experiment in which the sample was pumped at two frequencies simultaneously: $v_{p1} = 780$ MHz and $v_{p2} = 750$ MHz $< v_{p1}$. The static magnetic field was rotated by an angle of 45°, which made possible simultaneous parametric excitation of magnons by the spiral and coaxial resonators. The effect of the first pump on the limiting field of the second pump, i.e., actually the dependence of the NMR resonance field with frequency $v_{p2}/2$ on the power of the first pump, was studied. It was found that the NMR frequency still does not change even at the main pump power for which the shift of the AFMR line is already significant. For large supercriticalities, when $h/h_{c2} > 10$ and the maximum change in H_e corresponds to a decrease in the nuclear magnetization $\langle m \rangle$ by a factor ~ 1.5 , the shift of the NMR line corresponds to a decrease in $\langle m \rangle$ by only a factor 1.14 (Fig. 13, curves 4 and 5). This is another confirmation of the fact that the formula (5)(3) does not give the correct relation between the AFMR and NMR frequencies with large nuclear-magnon occupation numbers. We note that the effects studied here are not directly related to the hard character of the parametric excitation of magnons and that we also observed them at $\omega_{p1}/2\pi = 900$ MHz, i.e., with very low hardness.

Similar investigations of the effect of parametric excitation of nuclear magnons on the NMR frequency in $CsMnF_3$ were previously conducted in Ref. 16. The NMR frequency started to increase in a threshold manner when the critical pump field of the nuclear magnons was significantly exceeded, i.e., the NMR spectrum did not change up to some value of the amplitude of the parametric magnons. We note that the effect of parametric NSWs on both the AFMR and NMR spectra was not investigated in Refs. 15 and 16. Correspondingly, a comparative analysis of the shifts of the AFMR and NMR lines was also not performed.

Thus in the present work it is established for the first time that in the case when the nuclear magnetization decreases as a result of parametric excitation of high-amplitude NSWs and the nuclear subsystem is in a state of flow equilibrium and not thermodynamic equilibrium, the relation between the electronic and nuclear spin-wave spectra changes and is no longer described by the theory given in Ref. 1. This circumstance was neglected in many works, where the position of the AFMR line was used to determine the temperature and spectrum of a strongly excited nuclear system.

Our further investigations showed that parametric excitation at a frequency ω_{p1} not only shifts the AFMR and NMR lines, but also affects both the threshold h_{c1} and the supercritical behavior of the magnons excited by the second pumping of NSWs ($\omega_{p2} < \omega_{p1}$). The latter effect is manifested as a change in the amplitude of the modulational response α_{m2} of magnons with frequency $\omega_k = \omega_{p2}/2$. In addition, the "upper" pump for any frequencies ω_{p1} and ω_{p2} affects the "lower" pump. However, even the maximum power of the "upper" pump changes significantly the modulational response α_{m2} only when the second pump is not far above threshold, i.e., when the number of parametric magnons of frequency ω_{k2} in the sample does not exceed ~ 10¹⁴. It is interesting that in the case $\omega_{p1} < \omega_{p2}$ turning on the first pump has no effect on the amplitudes h_{c1} and α_{m2} of the second pump.

It has thus been established that "diffusion" of nuclear spin waves along the spectrum proceeds in the direction of decreasing frequencies and wave numbers, i.e., under conditions of parametric excitation of NSWs the secondary magnons are concentrated in the range from $\omega_p/2$ up to the nuclear magnetic resonance frequency ω_n .

DISCUSSION

We propose the following qualitative explanation of the disagreement between the experimental results and the theoretical spectra (3).

Each magnon has a magnetic moment which decreases the average nuclear magnetization. At high amplitude of parametric and secondary NSWs this decrease can be large, comparable to the nominal value of $\langle m \rangle$ at the given temperature, and this is why the AFMR frequency changes. Since most of the nuclear heat capacity is concentrated in the short-wavelength oscillations of the system, and pumping overheats a comparatively narrow region of the NSW spectrum ($\mathbf{k} < 10^5$ cm⁻¹), the nuclear temperature does not increase significantly. However, when the spin-wave amplitude is large the approximation of small oscillations, in which the frequencies of the electronic and nuclear magnons are calculated, does not work.

We estimated the number of parametric nuclear magnons and compared it with the nuclear magnetization at thermodynamic equilibrium. We obtained the following results.

1. Our 0.02 cm³ CsMnF₃ single crystal contained $2 \cdot 10^{20}$ Mn nuclei. The degree of polarization of the nuclear system at T = 1.55 K is equal to

$$\langle l \rangle = \frac{I(l+1)}{3} \frac{\hbar \omega_n}{k_B T} \approx 6 \cdot 10^{-2},$$

i.e., there are six nuclear magnetons $(6\mu_N)$ per 100 Mn nuclei, and the total magnetic moment of the nuclear system of the sample is equal to $10^{19}\mu_N$.

2. For supercriticality $h/h_{c2} \sim 10$ the number of parametric nuclear magnons is approximately equal to $5 \cdot 10^{15}$. The magnetic moment of NSWs is the sum of the contributions from the electronic and nuclear systems and is equal to $\sim 30\mu_N$. Multiplying this quantity by μ_N/μ_B we find that a single NSW decreases the nuclear magnetization by $\sim 10^{-2}\mu_N$, i.e., the parametric NSWs have a total magnetic moment $\sim 10^{14} \mu_N$.

It is obvious that the density of parametric NSWs is too low to change appreciably the average nuclear magnetization. However, the shift of the AFMR line indicates that the nuclear magnetization changes by a large amount. This inconsistency can be eliminated by assuming that the number of secondary NSWs is $\sim 10^4$ times larger than the number of parametric NSWs.

The number of secondary magnons can be roughly estimated by measuring the effect of the secondary pump magnons v_{p1} on the modulational response α_{m2} of the parametric magnons of the second pumping $v_{p2} < v_{p1}$. It can be conjectured that this effect should be manifested when the number of secondary NSWs of the first pumping in the region $v_{n2}/2 \pm \Gamma$ is comparable to the number $N \sim 10^{14}$ of parametric NSWs of the second pumping. If it is assumed that the secondary magnons are distributed uniformly over the frequencies of the NSW spectrum in the range from $v_{p1}/2$ up to v_n , then their total number is $N_s \sim 10^{18}$. Thus, according to this estimate, the number of secondary magnons of the first pumping is two orders of magnitude higher than the number of parametric magnons of this pumping, i.e., the number of secondary magnons is close to the number of polarized nuclear spins.

Obviously, significant accumulation of secondary magnons can occur if at the temperature $T \approx 1.5$ K energy flows out of the nuclear spin system within a 10^2-10^3 times longer time than the relaxation time of a parametric magnon $(\tau = 10 \,\mu\text{s})$. This means that the thermodynamic theory of parametric resonance¹⁷ should be applicable for the system of NSWs, since the coupling of the nuclear system with external systems is weak and energy is redistributed within the magnon system much more rapidly than energy is transferred into other degrees of freedom of the crystal.

We now attempt to explain on the basis of the foregoing analysis the long defect cooling times observed in Fig. 11. As the experiments show, for defect-cooling times $\tau \sim 1$ s the temperature of the nuclear system increases by a small amount, not exceeding 10%. Evidently, the observed anomalously high values of τ are not directly related to the thermodynamic overheating of the nuclear system. The experimental results clearly show that the defect relaxation time depends on the existence of nonequilibrium magnons in the sample. When the secondary-magnon occupation numbers increase significantly the temperature of the defects absorbing these magnons is higher than that of the lattice, until the energy of the secondary magnons is transferred from the nuclear system to the lattice. Then the relaxation time of the defects is determined by the relaxation of the number of secondary magnons.

As shown in Ref. 4, the relaxation rate of nuclear magnons in $CsMnF_3$ and $MnCO_3$ has the form

$$\Gamma_m = f_1 T + f_2 T^5,$$

where $f_{1,2}$ are functions of the magnon frequency and wave vector. The first term is governed by quasielastic scattering NSWs, i.e., it does not change the number of NSWs. The second term represents the sum of the magnon-phonon and three-ferromagnon relaxation processes (the three-ferromagnon process exists only in six-sublattice CsMnF₃). These relaxation processes reduce the number of NSWs, and for CsMnF₃ at T = 1.55 K and $v_k \approx 390$ MHz they give $f_2T^5 \sim 100$ Hz, which is almost 100 times lower than Γ_m . Corresponding estimates for the experimental conditions of Ref. 15 give $f_2T^5 \sim 160$ Hz. Nonetheless these values are still too high to describe the experimental results.

The situation is even worse when it comes to explaining $\tau_{\max} \sim 10^5 \,\mu$ s (Fig. 11) at T = 4.2 K. In this case $f_2 T^5 \sim 20$ kHz, i.e., the energy of a paramagnetic nuclear magnon is transferred to other degrees of freedom of the crystal in a time $\tau_m \sim 10 \,\mu$ s. This is four orders of magnitude higher than the maximum defect cooling time τ_{\max} . It can be conjectured that magnons are strongly accumulated at the bottom of the NSW band, since the secondary magnons diffuse in the direction of small k and the conservation laws forbid the magnon-phonon process for $\mathbf{k} < 10^4$ cm⁻¹. In this case, however, the magnons should affect strongly the relaxation time of the defects only if $\omega_n \approx \omega_{def}$. Thus, at the present time we do not have a qualitative model that describes adequately the experimentally observed increase in the defect relaxation time in the case of strongly excited NSWs.

As noted above, NSWs "diffuse" primarily in the direction of low frequencies. This result explains the unsymmetric shape of the lines for probe power $P_{\rm pr}$ with which the defects are saturated. For $\omega_{\rm pr} > \omega_p/2$ defects with frequency $\omega_{\rm pr}$ absorb energy from the probe pump, and then emit magnons with frequency $\omega_{\rm pr}$. These magnons diffuse downwards along the spectrum, and then some of them are absorbed by defects with frequency $\omega_p/2$, which decreases the parametric-excitation threshold h_{c1} . In the case $\omega_{\rm pr} < \omega_p/2$ the effect on h_{c1} is significantly smaller, since diffusion upwards along the spectrum is much weaker.

In conclusion we note that in spite of the large amount of experimental data available on the relaxation of electronic

and nuclear spin waves and defects, the nature of the defects which we have studied is still not fully understood. In Ref. 9 it was conjectured that the hardness of NSWs is due to the breakdown of the stoichiometry of the crystal, i.e., excess Mn atoms. In the present work we measured the hardness in several nonstoichiometric samples. We found that an excess of any element Mn, Cs, or F does not significantly affect either the frequency or the magnitude of the peak of negative nonlinear damping of NSWs. By surface grinding the sample we were able to approximately double the turning-off part of the relaxation; this could be associated with an increase in the number of dislocations in the crystal. Thus our results are consistent with Refs. 18–20, where the main defects affecting magnon relaxation in CsMnF₃ were conjectured to be dislocations.

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- ¹⁾ So-called elastic interactions of magnons result in gradual "diffusion" along the magnon spectrum, because the quasiparticle energy is determined to within the uncertainty relation. For this reason, it is more accurate to term these two-magnon processes quasielastic.
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