Investigation of the interaction between nonequilibrium phonons and nuclear spin waves in antiferromagnetic $CsMnF_3$

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An increase in the relaxation rate of the nuclear spin system in the easy-plane antiferromagnet $CsMnF_3$ during the excitation of phonons has been experimentally observed. It is shown that the nonequilibrium phonons make an additional contribution $\Delta\Gamma$ to the relaxation in the case in which their frequency corresponds to the spectral frequencies of the nuclear spin waves (NSW). The long-wave-NSW relaxation processes that can occur when the phonon subsystem of the antiferromagnet is in the nonequilibrium state are analyzed theoretically. It is shown that the main relaxation channel for the long-wave NSW is the scattering of these waves by the NSW excited by the nonequilibrium phonons. Two NSW excitation mechanisms are considered: the mechanism of linear conversion of a phonon into a NSW during the interaction with the fluctuations of the z component of the paramagnetic nuclear spin and the paramagnetic mechanism, in which two NSW are produced by two phonons. A comparison of the experimental and theoretical results shows that the observed characteristics of the behavior of $\Delta\Gamma$ can be explained within the framework of the proposed interaction mechanisms. The method used in the present investigation to speed up the nonequilibrium-phonon induced transverse-relaxation processes allowed the experimental separation of the α and β mechanisms of production of frequency-modulated (FM) echo.

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1. INTRODUCTION

A nuclear spin system with a dynamic frequency shift (DFS) is one of the most interesting objects that can be investigated by the NMR techniques, owing to a number of nonlinear properties possessed by these systems. The DFS is observed in a number of magnetically ordered crystals at sufficiently low temperatures under conditions when the magnetic-resonance frequencies of the electronic spin system are close to the NMR frequencies. The results are coupled long-wave oscillations of the electronic system, ordered by the exchange interaction, and of the paramagnetic system of nuclear spins. The quasinuclear mode of these oscillations possesses both the normal properties of paramagnetic systems (dependence of the magnitude of the nuclear magnetization on the temperature according to the Curie law, relatively long relaxation times) and properties stemming from the joint precession with the ordered electronic spin system. These properties can be divided into three groups:

1. There exist at the NMR frequencies well defined quasiparticles—nuclear spin waves (NSW), which are coupled vibrations of the paramagnetic nuclear spin system and the electronic spin system. The vibrational spectrum of the quasinuclear mode (called below, for simplicity, NSW) and the quasielectronic mode (hereafter called spin waves) of the antiferromagnet has the form[3)]

$$\omega_{nk}=\omega_n(1-(\mu H_\Delta)^2/\varepsilon_k^2)^{1/2},\quad \varepsilon_k=(\varepsilon_0^2+s^2k^2)^{1/2},\qquad (1)$$

where $\omega_n=\gamma H_n$ is the unshifted NMR frequency, $H_n=AM_0$ is the hyperfine field at the nuclei, $H_\Delta{}^2=2H_EH_N$, $H_N=A\langle m_z\rangle$ is the averaged field acting on the electrons, $H_E=\delta M_0$ is the exchange field, M_0 is the sublattice magnetization, ε_k is the spin-wave energy, and s is the maximum spin-wave velocity.

2. The uniform-precession frequency ω_{n0} of the nuclear spins is shifted with respect to the quantity ω_n as a result of the hyperfine field at the nuclei [see (1) with $k=0$]. This shift and the dispersion in the nuclear subsystem for $k\neq 0$ have a dynamical character, since they occur as a result of the precession of the electronic magnetization at the NMR frequency. In other words, the dispersion that arises in the nuclear subsystem is a consequence of the indirect, virtual-spin-wave-mediated interaction between the nuclear spins (the Suhl–Nakamura interaction).

3. The anisotropy in the properties of the ordered magnetic subsystem leads to a situation in which the resultant nuclear-electronic magnetization at the NMR frequencies moves in a noncircular orbit, and its component along the external magnetic field oscillates with, as a rule, a doubled frequency. Therefore, the quasinuclear vibrational mode can be excited parametrically by varying the external-magnetic-field strength at the doubled frequency.

It can be seen directly from (1) that, as the wave vector increases, the coupling weakens, the dispersion of the NSW decreases, and the NSW frequency approaches the unshifted-NMR frequency ω_n. When the wave vector of the electronic spin wave exceeds the value $k^*\approx a^{-1}(\omega_n/T)^{2/3}$, the relaxation frequency of the nuclear subsystem exceeds the magnitude of the DFS, as a result of which the spatial dispersion can be neglected. Therefore, the behavior of the corresponding states of the nuclear subsystem has an entirely paramagnetic character. This circumstance leads to important distinctive features in the shaping of the relaxation and kinetic processes involving the nuclear subsystem.

The magnitude of the DFS depends on the relative orientation of the electron and nuclear spins, and this leads to a significant nonlinearity in the vibrations in question, i.e., to the dependence of the precession frequency on the deviation amplitude of the spins. The above-considered properties of spin systems with DFS give rise to a number of new effects. The properties of spin systems with DFS are discussed in detail in Turov and Petrov's book.[1] The investigations of these systems by pulse methods are reviewed in Ref. 2; the investigations by acoustic NMR techniques, in Ref. 3. The properties of nuclear spin waves have been investigated by the methods of parametric pumping by both radio-frequency fields[4,5] and hypersound.[6,7]

The relaxation processes in nuclear systems with DFS have been studied in a number of experimental investigations.[5-9] It has been found[5b,8,9] that the temperature dependence of the relaxation time of the transverse component of the nuclear magnetization in the system of Mn^{55} nuclei in $MnCO_3$ has the form $T_2^{-1} \sim T^{5-6}$ at $T > T_0 = 3.5$ K and the form $T_2^{-1} \sim T$ for $T < T_0$. One of the present authors (Yu. M. B) has observed a similar behavior of the relaxation time T_2 in $CsMnF_3$, for which the characteristic temperature $T_0 \approx 3.8$ K. A theoretical analysis of the relaxation mechanisms for long-wave NSW has shown[10] that, for NSW with $k < \varepsilon_0/s$, the dominant relaxation process in the $T < T_0$ temperature region is the NSW–NSW scattering process stemming from the spin–spin interaction. In the temperature region $T > T_0$ the interaction of the NSW with the equilibrium phonons, which makes a contribution $\sim T^5$, is important.

The present paper is devoted to the theoretical and experimental investigation of the effect of the nonequilibrium condition of the phonon subsystem on the relaxation properties of a nuclear spin system with a DFS.

2. MEASUREMENT PROCEDURE

For the investigations we chose the antiferromagnet $CsMnF_3$, which possesses the "easy-plane" type of anisotropy, and whose magnetic properties have been well studied. The low-frequency part of the spectrum of the magnetic and phonon vibrations in $CsMnF_3$ is shown in Fig. 1. The existence of two NSW branches

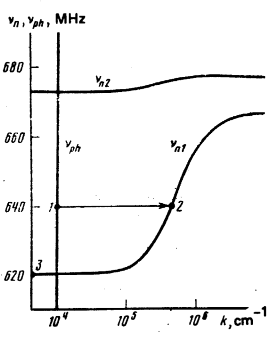

FIG. 1. Low-frequency part of the spectrum of the magnetic and phonon vibrations in $CsMnF_3$. Nonequilibrium phonons excited at the point 1 excite NSW at the point 2, which in turn speed up the transverse relaxation of the uniform precession (3). The spectrum was constructed from the data given in Refs. 11–13.

with DFS is due to the fact that the magnetic unit cell of $CsMnF_3$ contains six Mn^{++} ions located at two inequivalent sites. The hyperfine fields acting on the Mn^{55} nuclei of these ions are equal to 666 and 678 MHz (in frequency units). The low-frequency NSW branch corresponds to the in-phase motions of the nuclear subsystems of the two ions, while the branch lying in the frequency interval from 666 to 678 MHz corresponds to the antiphased motions of the nuclear spins of the four ions of one site, on the one hand, and the nuclear spins of the two ions of the other site, on the other.

The investigations were performed on a synthetic $CsMnF_3$ single crystal grown at our Institute.[4)] The sample had the shape of a cylinder with diameter = 4 mm and length $l = 3$ mm, with its axis lying in the plane of easy magnetization. One of the ends of the sample had an optically polished surface. This end was pressed against a $LiNbO_3$-single-crystal cylinder, which was used as a phonon generator.

The investigations were performed with a pulse spectrometer, a block diagram of which is shown in Fig. 2. The acoustic part of the spectrometer consists of a GCh-37A oscillator, a variable attenuator, and a circulator, matching lines and a plane-parallel capacitor, and a RF electric field that excited phonons in the $LiNbO_3$. The design of the acoustic transducer is similar to that of the transducer described in Ref. 14. The acoustic-response signal of the $LiNbO_3$ crystal could be fed to a P5-9 superheterodyne receiver, and observed on an oscillograph. This signal was used to tune the acoustic part of the spectrometer. To obtain a higher RF radiation power, the oscillator was modulated by feeding a pulsed anode voltage to the generating tube. The RF radiation power than attained a value of 10 W. If we assume the efficiency of the transducer and the RF circuit to be equal to 0.1, then the energy flux density in the sound pulse is 10 W/cm^2.

The magnetic part of the spectrometer was a paramagnetic-echo spectrometer,[15] and consisted of a resonance-frequency generator, a doubled-frequency generator, attenuators, and matching lines. The RF magnetic fields of resonance and doubled frequencies were excited by a single-turn coil, inside which was the sample under investigation. The RF magnetic fields and the constant magnetic field lay in the plane of easy magnetization of the sample at an angle of 60° to each

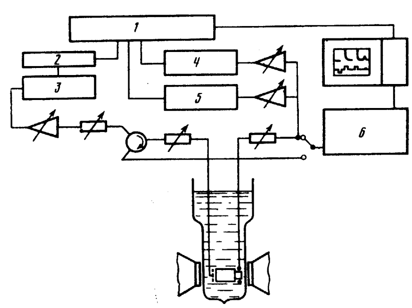

FIG. 2. Block diagram of spectrometer: 1) pulse supply unit, 2) modulator, 3) ω_{ph} generator, 4) ω_n generator, 5) $2\omega_n$ generator, and 6) receiver.

other. Thus, the RF fields had components directed both at right angles to, and along, the constant magnetic field. The RF magnetic field was polarized along the direction of propagation of the longitudinal phonons in the sample. The RF-magnetic-response signal from the sample was picked up by the same single-turn coil, and fed through the P5-9 superheterodyne receiver to the oscillograph.

The investigations were performed at NMR frequencies in the range from 500 to 700 MHz in fields of intensity ranging from 500 to 5000 Oe at temperatures in the range from 1.5 to 4.2 K.

3. EXPERIMENTAL INVESTIGATIONS OF THE EFFECT OF ACOUSTIC PUMPING ON THE RELAXATION PROCESSES

The transverse-relaxation rate of the uniform precession of the quasinuclear vibrational branch was measured, using the parametric-echo method,[15] which is applicable to resonance systems that admit of both resonance and parametric excitation. In this method the spin system is excited by a resonance RF pulse and, after a time lag t_{12}, by a parametric-pumping pulse (a RF-field pulse of doubled frequency with polarization parallel to the constant magnetic field). At the moment of time $2t_{12}$ the spin system's transverse magnetization produced by the first resonance pulse is partially restored, and this leads to the appearance in the receiving system of a RF signal—a parametric-echo signal. The mechanism underlying the formation the parametric echo differs from the earlier-known mechanisms underlying echo-signal production by linear properties, and this enables us to produce intense signals at small angles of deviation of the spins, as well as to easily interpret the results obtained. These properties of the parametric echo are quite important for the investigation of systems with coupled nuclear-electronic precessions, since instability processes develop in them at high excitation amplitudes. Furthermore, the production of a spin echo during the excitation of the system in question by the two resonance pulses occurs as a result of a change in the eigenfrequencies of the spins—the frequency modulation (FM) mechanism. The intensity of the FM-echo signal generated in this case depends complexly on the time lag between the pulses, which makes the use of this method for the investigation of the relaxation processes difficult. In contrast, the dependence of the intensity of the parametric echo on the time lag between the pulses is entirely determined by the relaxation processes, and has the form

$$I \sim \exp(-2t_{12}/T_2).$$

We found that the presence in the sample of nonequilibrium phonons at NSW frequencies leads to the acceleration of the decay of the parametric-echo signal, an observation which we interpret as the shortening of the transverse-relaxation time of the uniform precession of the NSW. Figure 3 shows the dependence of the intensity of the parametric-echo signal on the time lag under equilibrium conditions and in the case in which an acoustic phonon-pumping pulse was fed to the sample just before the first resonance RF pulse. The nonequilibrium phonons do not lead directly to the heating of the spin system, and do not change the coupling between the nuclear and electronic precessions. This can be seen from the fact that the echo-signal intensities in Fig. 3 become equal when extrapolated to $t_{12} = 0$. Moreover, we found on checking that the NMR signal did not change its frequency after the supply of the phonon-pumping pulse.

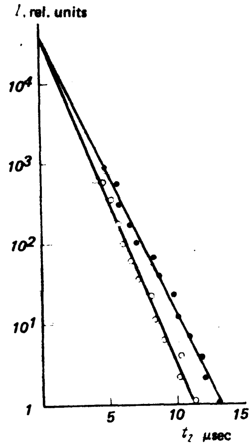

FIG. 3. The decay of the parameteric-echo signal in the case in which an acoustic-pumping pulse is supplied (○) and in the case in which such a pulse is not supplied (●).

It is convenient to describe the change in the transverse relaxation rate of the uniform precession in terms of the homogeneous resonance-line broadening $\Gamma = T_2^{-1}$. The pumping of nonequilibrium phonons leads to a change, $\Delta\Gamma$, in the broadening, which was obtained in the subsequent experiments from the results of measurements of the change in the transverse-relaxation rate. The phonon-pumping pulse speeds up the relaxation process not only at the time of its supply, but also for some time after that. We measured the quantity $\Delta\Gamma$ as a function of the time lag t_{ph} between the phonon-pumping pulse and the first of the RF-pulse train producing the spin echo. It turned out that the quantity $\Delta\Gamma$ decreases with increasing t_{ph} according to an exponential law with a time constant T_{ph} equal to 26 μsec at 1.5 K, 19 μsec at 2 K, and 11 μsec at 4.2 K.

Under the experimental conditions the time constant of the transverse relaxation of the uniform precession of the nuclear magnetization at 4.2 K was equal to $T_2 = 4$ μsec, i.e., it was significantly less than T_{ph}. Therefore, the contribution of the nonequilibrium phonons to the relaxation was practically time-independent over a measurement time of T_2. At lower temperatures T_2 approached T_{ph}, and therefore it was necessary to take into account the variation of $\Delta\Gamma$ during the formation of the spin-echo signal.

Besides the relaxation effects, the variation of the echo-signal intensity during the action of the phonon-pumping pulse could, in principle, be related to the fluctuations in the local fields at the nuclei. But in this case the effect would not have had a strongly pronounced frequency dependence. In our case a distinctive de-

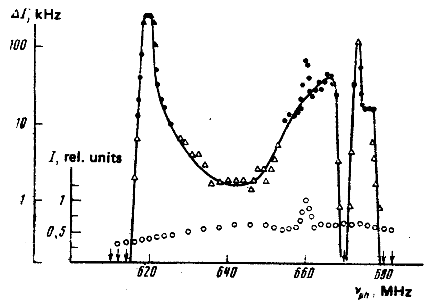

FIG. 4. Increase in the homogeneous broadening of the NMR line ($\Delta\Gamma$) as a result of the action of an acoustic-pumping pulse of frequency ν_{ph} (△, ▲, ●), and the operational efficiency of the acoustic transducer (○).

pendence of the quantity $\Delta\Gamma$ on the frequency of the pumped phonons is observed. Figure 4 shows the dependence of $\Delta\Gamma$ on the frequency of the nonequilibrium phonons at 4.2 K and in an external field such that $\nu_{n0} = \omega_{n0}/2\pi = 620$ MHz.

The measurements whose results are indicated in Fig. 4 by the points were performed with phonon-pumping pulses of duration 2 μsec. In the region of the strongly pronounced peaks at ν_{ph} equal to 620 and 673 MHz, the sensitivity of the spin system to the influence of the nonequilibrium-phonon pulse turned out to be so high that it was difficult to observe the parametric-echo signal in a fairly broad range of values of the time lag t_{12}. Therefore, the phonon-pumping pulse length was decreased to 1.5 μsec, and the measured $\Delta\Gamma$ values were multiplied by 1.77; the resulting values are indicated in Fig. 4 by the black triangles. The factor 1.77 was taken in accordance with the dependence $\Delta\Gamma \sim \tau_{ph}^2$, which was established experimentally, and which is discussed below. A similar procedure was adopted when the observed acceleration of the relaxation process due to 2-μsec pulses turned out to be very low. The open triangles in Fig. 4 indicate the $\Delta\Gamma$ values obtained with 5-μsec phonon-pumping pulses and then multiplied by 0.16. The arrows on the graph indicate the frequencies at which the effect of the phonon-pumping pulse on the relaxation processes was not detectable even when the pulse length was 6 μsec and the RF-radiation power fed to the electroacoustic transducer was 10 W. If we assume that the dependences $\Delta\Gamma \sim \tau_{ph}^2$ and $\Delta\Gamma \sim P^2$ obtain in this region, then in the case of the phonon-pumping pulse for which the graph in Fig. 4 was constructed the magnitude of $\Delta\Gamma$ should be less than 0.2 kHz at these points.

It is worth noting that the nonequilibrium phonons speed up the transverse uniform-precession relaxation process only when their frequencies correspond to the frequencies of the NSW spectrum, the peaks of this dependence being observed at 620, 673 MHz ($k=0$) and 666, 676 MHz ($k \approx k^*$). Figure 4 qualitatively corresponds to the frequency dependence of the density of NSW states in the region of phonon frequencies close to the unshifted-resonance frequency. The peak observed in the experimental dependence at $\nu^* = 660$ MHz is connected with the resonance properties of the acoustic circuit of the spectrometer, and is clearly visible in

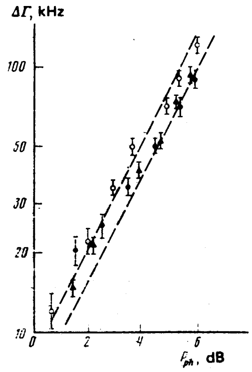

FIG. 5. Dependence of the increase ($\Delta\Gamma$) in the homogeneous broadening of the NMR line on the acoustic-pumping pulse power at frequencies ν_{ph} equal to 620 MHz (○), 666 MHz (●), and 676 MHz (▲) in cases of pulse durations respectively equal to 2, 4, and 4 μsec.

the case in which the acoustic-response signal is used to tune the spectrometer (the circles in the figure).

A qualitative analysis of the temperature dependence of the quantity $\Delta\Gamma$ in the case in which the external magnetic field is varied simultaneously with the temperature in such a way that the frequency ν_{n0} remains constant was carried out. It was found that the magnitude of $\Delta\Gamma$ decreases with decreasing temperature, with $\Delta\Gamma \sim T^2 - T^3$.

The dependence of $\Delta\Gamma$ on the power P and duration τ_{ph} of the phonon pulse was investigated at pumped-phonon frequencies of 620, 666, and 676 MHz (Figs. 5 and 6). It can be inferred from the experimental data that $\Delta\Gamma \sim P^2$ and $\Delta\Gamma \sim \tau_{ph}^2$, i.e., the additional relaxation is proportional to the square of the total number of nonequilibrium phonons. Thus, the participation of phonons in the transverse uniform-precession relaxation process occurring in systems with DFS of the NMR has been experimentally observed.

4. INVESTIGATION OF THE FM MECHANISM OF ECHO PRODUCTION

As noted above, in systems with DFS the spin-echo signal produced after two resonance RF pulses is form-

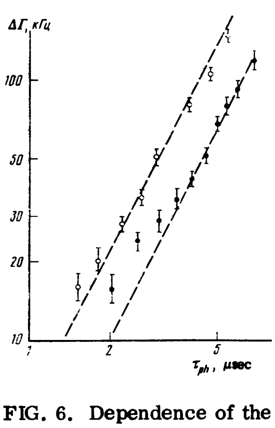

FIG. 6. Dependence of the increase in the homogeneous broadening of the NMR line on the acoustic-pumping pulse duration at frequencies ν_{ph} equal to 620 MHz (○) and 666 MHz (●).

ed by the frequency mechanism of echo production, a mechanism which has been actively investigated in recent years.[2] The FM echo arises as a result of the modulation of the distribution of the number of spins over the frequencies on account of the dependence of the spin-precession frequency on the angle of deviation of the nuclear magnetization from the equilibrium direction. Here the mechanism responsible for the forming of the FM echo is often divided into α and β mechanisms. In the α mechanism the second RF pulse phases, as it were, the transverse nuclear magnetization produced after the first pulse. In the β mechanism the echo signal is formed from the transverse magnetization produced by the second RF pulse. As a result, for the α mechanism the spin–spin relaxation process is switched on at the instant the first pulse begins to act; for the β mechanism, at the instant the second pulse begins to act. The concept of α and β mechanisms of echo production was first introduced[16] in 1973. But it has thus far not been possible to experimentally separate the contributions of the two mechanisms of echo production. Using the phonon-pumping method, we have succeeded in qualitatively separating the contributions of the α and β mechanisms to the generation of the FM echo.

To the sample were fed two resonance RF pulses with a time lag of 20 μsec between them. A FM-echo signal was observed at the instant $t=40$ μsec. In addition, a phonon-pumping pulse was fed to the system. The phonon pulse then effectively speeded up the transverse relaxation process only for a time $\sim T_{ph}$ after it. Under the conditions of the experiment $T_{ph}=19$ μsec $\approx t_{12}$, which allowed us to effectively speed up the relaxation process at different stages of the generation of the FM-echo signal by feeding the phonon-pumping pulse at different times.

Figure 7 shows the dependence of the intensity of the FM-echo signal on the feeding time of the phonon-pumping pulse. The rectangles indicate the feeding times of the RF pulses. In the case in which the relaxation process involving the transverse magnetization components generating the echo signal begins immediately after the first RF pulse (as obtains for the α mechanism, as well as in the classical Hahn echo and the parametric echo), the effect of the phonon pulse is greatest when $t_{ph}=0$. Indeed, there is time, when $t_{ph} > 0$, for the transverse relaxation rate to get partially restored by the time the first pulse is supplied. When

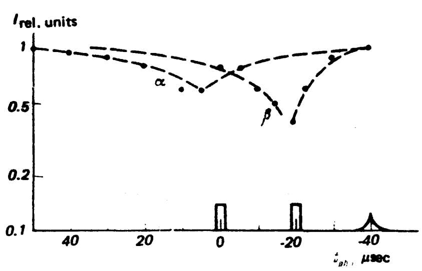

FIG. 7. Dependence of the intensity of the FM-echo signal on the time at which the acoustic-pumping pulse is supplied.

$t_{ph} < 0$, the spins relax at the normal rate in a time interval equal to t_{ph}. The first minimum of the echo-signal intensity in Fig. 7 (i.e., the minimum for $t_{ph}=3$ μsec) is due to the existence of the α mechanism of FM-echo production. The shift from the point $t_{ph}=0$ occurs because of the fact that the phonon pulse takes 3 μsec to travel through the $LiNbO_3$ crystal. A second spin-echo-intensity minimum is clearly visible at $t_{ph}=t_2+3$ μsec, which shows that part of the transverse magnetization producing the echo signal begins to relax after the second RF pulse, as should be the case for the β mechanism of FM-echo production, the β mechanism making the dominant contribution to the intensity of the echo signal under the conditions of our experiment.

5. THE MECHANISMS UNDERLYING THE INFLUENCE OF PHONONS ON THE RELAXATION OF NSW

As has already been noted in the Introduction, the behavior of the nuclear subsystem in magnetically ordered materials at low temperatures has a number of characteristics: it has a wave character in the long-wave region, whereas the short-wave states are entirely paramagnetic. In view of this, the description of nuclear subsystems with DFS in the spin representation (in particular, the use of the diagrammatic technique for the spin operators) seems to be adequate. The indicated specific properties of the systems in question can, however, be taken into consideration with the aid of a previously proposed approach.[10] In this approach the phase space of the nuclear subsystem is divided into two regions: the long-wave ($k < k^*$) and the short-wave ($k > k^*$) region. In the long-wave region the oscillations of the nuclear magnetization are described by excitations of the Bose type, while the short-wave states are considered in terms of quantum transitions between paramagnetic levels.

Let us write the Hamiltonian in the form

$$\mathscr{H}=\mathscr{H}_{m-n}+\mathscr{H}_{m-ph},$$

where

$$\mathscr{H}_{m-n}=\int dr\left[\frac{\delta}{2}M^2+\frac{b}{2}L_z^2+\frac{\alpha}{2}\frac{\partial \mathbf{L}}{\partial x_i}\frac{\partial \mathbf{L}}{\partial x_i}+d(L_xM_y-L_yM_x)-\mathbf{MH}+\frac{A}{2}(\mathbf{Ll}+\mathbf{Mm})\right] \quad (2)$$

is the Hamiltonian of the electronic and nuclear subsystems in the two-sublattice model[5]; M, L and m, l are respectively the ferromagnetic- and antiferromagnetic-moment vectors for the two subsystems; δ is the homogeneous-exchange parameter; b is the anisotropy constant; α is the inhomogeneous-exchange parameter; d is the Dzaloshinskiĭ constant; H is the external magnetic field;

$$\mathscr{H}_{m-ph}=\int dr\{B_{12}[(L_x^2-L_y^2)(u_{xx}-u_{yy})+4L_xL_yu_{xy}]+B_{14}[(L_x^2-L_y^2)u_{yz}+2L_xL_yu_{xz}]\} \quad (3)$$

is the magnetoelastic-interaction Hamiltonian for a rhombohedral antiferromagnet, in which only the terms describing the interaction of the phonons with the low-frequency branch of the spin waves are given; B_{12} and B_{14} are magnetoelastic coupling constants; and $u_{\alpha\beta}$ is the elastic-strain tensor. We have, for the sake of generality, included in (2) the term responsible for weak ferromagnetism, and $\mathscr{H}_{m\to ph}$ has been written for crystals of rhombohedral symmetry. To apply it to hexagonal $CsMnF_3$, we should set $d=0$ and $B_{14}=0$.

The transition from the quantities **M**, **L** and **m**, **l** in the long-wave region to the spin-deviation operators is carried out in standard fashion. The diagonalization of the resulting quadratic form determines the spectrum of the spin waves and the nuclear spin waves. The explicit form of the low-frequency type spectrum of these vibrations is given by the formula (1). The expansion of the last term in (2) in the long-wave ($k_i < k^*$) region to fourth order in the Bose amplitudes, with the subsequent transition to the normal-mode amplitudes reveals four-wave anharmonicities in which the nuclear spin waves participate.[10]

In accordance with the fact that the phase space of the nuclear subsystem has two characteristic regions (the long-wave and the short-wave), there exist, besides the interaction processes in the long-wave NSW system, interaction processes involving the long-wave quasiparticles and the entirely paramagnetic states of the nuclear subsystem. In the present paper we shall be interested in the interaction of the waves with the fluctuations in the z component of the paramagnetic nuclear spin. The effective Hamiltonian of processes of this type is given in the Appendix [see (A. I. 1)].

By going over in the magnetoelastic Hamiltonian (3) from the elastic-displacement vector to the phonon creation, ($b_{q\lambda}^+$) and annihilation ($b_{q\lambda}$) operators and from the antiferromagnetism vector **L** to the creation and annihilation operators for the low-activation spin waves, we can easily take into account the linear coupling between the SW and the phonons, as well as the three-wave interaction processes involving the SW and the phonons.[17] Allowance for the dynamical linear coupling between the electronic and nuclear subsystems enables us to obtain the linear NSW–phonon coupling responsible for the nuclear magnetoacoustic resonance and the three-wave interaction processes involving the NSW, the SW, and the phonons.[10]

It should be noted that all the three-wave interactions of the NSW with the spin waves and the phonons are forbidden by the conservation laws at phonon frequencies of $\Omega \approx \omega_n$. The conservation laws may allow the NSW–phonon interaction processes in which four quasiparticles participate. Some of these processes are depicted in Fig. 8(a). The amplitudes of processes of this type arise from both the three-wave interaction processes with the participation of spin waves, nuclear spin waves, and phonons in second-order perturbation theory and the four-wave processes with allowance for the linear phonon–spin wave and phonon–NSW couplings [see Fig. 8(b) and the Appendix II]. The direct contribution to the NSW relaxation of the four-wave interaction processes in which the nonequilibrium phonons participate is given by the following relation:

$$\Delta\Gamma \sim \frac{\Theta^2}{Mv^2}\left(\frac{\omega_n}{\varepsilon_0}\right)^3 \frac{T}{\varepsilon_0}\left(\frac{J_0}{\omega_D}\right)^3 N_{ph};$$

for the characteristic parameter values (see below) it is small: $\Delta\Gamma < 10^{-2}$ Hz.

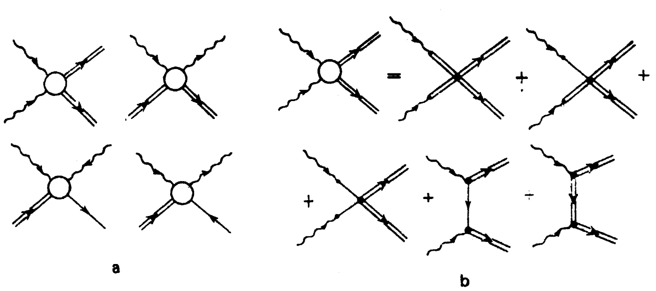

FIG. 8. a) Four-particle processes of interaction of NSW (double line) with phonons (wavy line) and magnons (solid line); b) diagrammatic representation of the contributions of the various mechanisms to the amplitude of the conversion of two phonons into two NSW.

Besides the nonlinear phonon–NSW interaction processes of a wave nature, there exist processes involving the linear conversion of phonons into NSW during the interaction with the fluctuations of the z component of the nuclear spin (see Fig. 9 and the Appendix III). These processes do not conserve the quasiparticle momenta, since the fluctuations corresponding to the short-wave part of the phase volume of the nuclear subsystem are not correlated in the space (there is no dispersion).

Since, as has been shown, the contribution of the direct NSW–nonequilibrium phonon interaction processes to the relaxation, $\Delta\Gamma$, of the uniform precession of the nuclear spins is negligible, let us consider a more complex mechanism through which the nonequilibrium phonons exert their influence on $\Delta\Gamma$. In this mechanism the nonequilibrium phonons cause the excitation of NSW with $k \neq 0$, which scatter the vibrational energy of the nuclear precession. As the mechanisms underlying the excitation of NSW by sound, we consider the following two mechanisms: 1) the mechanism of linear conversion of a phonon into a NSW by the fluctuations of the z component of the nuclear spin and 2) the parametric mechanism resulting from a nonlinear process of the type $ph + ph \to n + n$, and realized at a fairly high sound-pulse power.

Thus, let us consider the following terms in the effective NSW–phonon interaction potential under conditions when the phonon subsystem is in the nonequilibrium state:

$$\mathscr{H} = \sum_{\mathbf{k}} \omega_{n\mathbf{k}} c_{n\mathbf{k}}^+ c_{n\mathbf{k}} + \sum_{\mathbf{q}\lambda} \Omega_{\mathbf{q}\lambda} b_{\mathbf{q}\lambda}^+ b_{\mathbf{q}\lambda} + \frac{1}{N}\sum_{\mathbf{k},\mathbf{q},\lambda} [\hat{U}^z_{ph-n} b_{\mathbf{q}\lambda} c_{n\mathbf{k}}^+ + \text{H. c.}]$$
$$+ N^{-1/2} \sum_{\mathbf{k}_1+\mathbf{k}_2=\mathbf{q}} (\Psi_{2n-ph} b_{\mathbf{q}\lambda} \overset{+}{c}_{n\mathbf{k}_1} \overset{+}{c}_{n\mathbf{k}_2} + \text{H. c.}) \qquad (4)$$
$$+ N^{-1} \sum_{1+2=3+4} [\Phi_{2n-2ph} b_3 b_4 c_{n1}^+ c_{n2}^+ + \Phi_{4n} c_{n1}^+ c_{n2}^+ c_{n3} c_{n4} + \text{H. c.}],$$

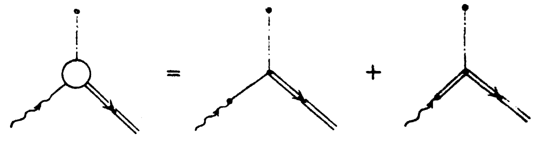

FIG. 9. Diagrammatic representation of the linear conversion of a phonon into a NSW on the fluctuations of the z component of a paramagnetic nuclear spin.

where $\Omega_{q\lambda}=v_\lambda q$ is the phonon frequency, v_λ is the velocity of sound,

$$\Psi_{2n-ph}=\Theta\left(\frac{\Omega_{q\lambda}}{2Mv^2}\right)^{1/2}\frac{J_0}{(\omega_{nk_1}\omega_{nk_2})^{1/2}}\frac{\omega_n^2(\mu H_\Delta)^2}{\varepsilon_{k_1}^2\varepsilon_{k_2}^2}\varphi(\mathbf{e},\mathbf{n}) \quad (5)$$

is the amplitude of the interaction of two NSW with a phonon, $\varphi(\mathbf{e},\mathbf{n})$ is a function of the order of unity [see (A.II.4a)], and

$$\Phi_{4n}=-\frac{J_0\omega_n^2}{32}\left(\frac{1}{\varepsilon_{k_1}^2}+\frac{1}{\varepsilon_{k_2}^2}+\frac{1}{\varepsilon_{k_3}^2}+\frac{1}{\varepsilon_{k_4}^2}\right) \quad (6)$$

is the NSW–NSW scattering amplitude. The explicit form of the quantities $\hat{U}^z_{ph-n}$ and Φ_{2n-2ph} is given in the Appendix. The factors J_0 and $H_\Delta^2=2H_EH_N$ in the expressions (5) and (6) indicate the manifestation of the effect of exchange strengthening of the magnetoelastic[18,19] and hyperfine[20,21] interactions in antiferromagnets with an easy-plane type of anisotropy.

We have introduced the following notation: $\Theta=B_{12}L_0^2V_0$ is the magnetoelastic energy; $L_0=2M_0$ is the antiferromagnetic vector; $M=\rho V_0$ is the mass, and V_0 the volume, of the unit cell; $J_0=\mu H_E/S$ is the intersublattice exchange parameter in energy units; S is the spin of the magnetic ion ($\frac{5}{2}$ for Mn^{55}); $\mathbf{e}$ is the phonon-polarization vector; and $\mathbf{n}=\mathbf{q}/|\mathbf{q}|$ is the unit vector along the direction of propagation of the phonon. The quantity V_0 used by us in the two-sublattice model is connected with the magnetization M_0 by the following relation: $M_0=\mu S/V_0$.

In the expression (4) the third term is responsible for the linear conversion of sound into NSW, the fourth and fifth lead to the parametric excitation of NSW by phonons, and the fourth and last terms determine the stationary level of parametrically excited NSW, the last term describing the interaction between the uniform precession of the nuclear spins and the nonequilibrium NSW with $k\neq 0$.

Let us first discuss the linear-conversion mechanism. This process, being a two-particle process, conserves the frequency, but not, as noted above, the wave vector, just like what happens in the scattering of waves by inhomogeneities. The kinetic equation for the occupation numbers $n_\mathbf{k}$ of the nuclear spin waves with allowance for the conversion of the phonons into NSW during their interaction with the fluctuations of the longitudinal component of the nuclear spin has the form[6)]

$$\frac{dn_\mathbf{k}}{dt}=2\pi V_0\int\frac{d\mathbf{q}}{(2\pi)^3}\langle|\hat{U}^z_{ph-n}|^2\rangle(N_\mathbf{q}-n_\mathbf{k})\delta(\Gamma_n|\Omega_\mathbf{q}-\omega_{n\mathbf{k}})+\mathscr{L}_n^{int},$$
$$\delta(\Gamma_n|\Omega_\mathbf{q}-\omega_{n\mathbf{k}})=\frac{1}{\pi}\frac{\Gamma_n(\mathbf{k})}{(\Omega_\mathbf{q}-\omega_{n\mathbf{k}})^2+\Gamma_n^2(\mathbf{k})}, \quad (7)$$

where the angle brackets indicate averaging over the nuclear-spin orientations, $\mathscr{L}_n^{int}$ describes the interaction of the NSW with the thermostat, $\Gamma_n=\frac{1}{2}\tau_n^{-1}$, τ_n is the NSW relaxation time, and the $N_\mathbf{q}$ are the occupation numbers of the phonons. By writing $\mathscr{L}_n^{int}$ in the $\tau_\mathbf{k}$ approximation, we can transform the expression (7) into the form

$$\dot{n}_\mathbf{k}=2\pi\langle|\hat{U}_{ph-n}|^2\rangle\,\delta(\Gamma_n|\Omega_\mathbf{q}-\omega_{n\mathbf{k}})N_{ph}-n_\mathbf{k}/\tau_n(\mathbf{k})$$

or, equivalently,

$$\dot{N}_n=KN_{ph}-N_n/\tau_n,$$

where

$$K=2\pi V_0\int\frac{d\mathbf{k}}{(2\pi)^3}\langle|\hat{U}^z_{ph-n}|^2\rangle\delta(\Gamma_n|\Omega_\mathbf{q}-\omega_{n\mathbf{k}})$$

is the coefficient of conversion of sound into NSW, τ_n is the total relaxation time for all the interaction processes, including the process of linear conversion of a NSW into a phonon, and

$$N_{ph}=V_0\int\frac{d\mathbf{q}}{(2\pi)^3}N(\Omega_\mathbf{q}),\quad N_n=V_0\int\frac{d\mathbf{k}}{(2\pi)^3}n_\mathbf{k}$$

are the phonon and NSW occupation numbers per unit cell.

We obtain for the steady[7)] state within the framework of the linear sound → NSW conversion mechanism under consideration the expression

$$N_n=K\tau_nN_{ph}, \quad (8)$$

which, after being multiplied by $\Omega_\mathbf{q}=\omega_{n\mathbf{k}}$, has the meaning of energy balance.

It should be noted that if the populations of the phonon states are not maintained at the steady-state level, then the quantity K has the meaning of a sound attenuation factor. A direct computation yields

$$K=\frac{1}{\pi}\langle|\hat{U}^z_{ph-n}|^2\rangle k^2V_0\Big/\frac{\partial\omega_{n\mathbf{k}}}{\partial k}. \quad (9)$$

Here the magnitude, k, of the wave vector is determined by the condition $\omega_{n\mathbf{k}}=\Omega_\mathbf{q}$.

The other mechanism of NSW excitation is the parametric-excitation mechanism, which stems from the nonlinear interaction between the NSW and the phonons. The parametric instability resulting from the decay of a phonon into two NSW was theoretically considered earlier[6] for the spin-flop phase of a cubic antiferromagnet, and has been experimentally observed.[7] Notice that the expression (5) allows us to obtain a threshold formula for the two-NSW decay of hypersound in rhombohedral antiferromagnets for an arbitrary direction of propagation of the acoustic wave. In the case of relatively low frequencies, which is the case of interest to us here, when the indicated process is forbidden by the conservation laws, the process of parametric production of two NSW by two phonons, to which the fourth term in the expression (4) corresponds, can be realized. In the case in which the phonon wave vector q_0 is significantly smaller than the NSW vector, such an instability is the acoustic analog of the second-order Suhl instability.

Knowing the amplitude for the conversion of two phonons into two NSW, we can derive in the usual manner an expression for the threshold phonon number in the second-order process:

$$N_{ph}^c=(\Gamma_n(\mathbf{k}_1)\Gamma_n(\mathbf{k}_2))^{1/2}/2|\Phi_{2n-2ph}|. \quad (10)$$

Expressing the phonon number per unit cell, N_{ph}, in terms of the strain amplitude in the wave, we obtain for the critical acoustic-wave energy flux far from the magnetoacoustic resonance the expression

$$P_c=C^{(2)}v\frac{\Gamma_n(\mathbf{k})}{\omega_n}\frac{\varepsilon_0^4}{\Theta^2J_0^2}\frac{\Omega-\omega_{n0}}{\omega_n}\frac{2}{3}\left[1+\left(\frac{\mu H_\Delta}{\varepsilon_0}\right)^2\frac{\omega_n}{\Omega-\omega_{n0}}\left(1+\frac{\varepsilon_0^2}{\varepsilon_k^2}\right)\right]^{-1}. \tag{11}$$

Here we take into account the fact that for the acoustic model of interest to us

$$\varphi'(\mathbf{e},\mathbf{n})=e_xn_y+e_yn_x=\sin{}^2/_3\pi=\sqrt{3}/2,$$

[see (A.II.4b)].

The occupation numbers of NSW with energies $\omega_{nk}=\Omega$ increase sharply at sound-pulse powers higher than the critical power. Their steady-state level (see footnote 7) is determined by one or another limiting mechanism. As mechanisms underlying the limitation of the nonlinear NSW-excitation level, we shall consider the following two mechanisms: the phase mechanism,[22] for which the last term in (4) is responsible, and the nonlinear-damping mechanism.

In the case of the phase mechanism, we have for the integral number, N_n, of nonequilibrium NSW per unit cell the expression

$$N_n=\frac{\Gamma_n(\mathbf{k})}{4|\Phi_{4n}(\mathbf{k},\mathbf{k})|}\left(\frac{P}{P_c}-1\right)^{1/2}=2\frac{\Gamma_n(\mathbf{k})}{\omega_n}\frac{\varepsilon_k^2}{\omega_nJ_0}\left(\frac{P}{P_c}-1\right)^{1/2}. \tag{12}$$

At relatively low acoustic-power supercriticalities, the nonlinear-damping mechanism may be important in determining the nonequilibrium-NSW level. Leading to the positive nonlinear damping of the NSW are the processes involving the coalescence of two parametric NSW into a phonon [see the third term in the expression (4)]. In this case

$$N_n=\frac{\Gamma_n(\mathbf{k})}{\eta}\left(\frac{P}{P_c}-1\right) \tag{13}$$

where η is the nonlinear-attenuation factor, given by the relation

$$\Gamma_n(\mathbf{k})=\Gamma_n^{(0)}(\mathbf{k})+\eta N_n.$$

It can be seen from the relations (8), (12), and (13) that the number of nonequilibrium NSW in both excitation mechanisms depends on the nuclear spin wave lifetime τ_n and consequently, on temperature. NSW relaxation in a crystal in the equilibrium state is considered in Ref. 10, where it is shown that the dominant NSW-relaxation mechanism in the region of wave numbers $k>\varepsilon_0/s$ is the process, described by the second term in the expression (A.I.1), of elastic scattering by the fluctuations of the z component of the nuclear spin (see also Ref. 23):

$$\Gamma_{2n}=\frac{\omega_n}{8\pi}\frac{skTJ_0}{\Theta_N^3}, \tag{14}$$

where $\Theta_N=s/V_0^{1/3}$ and T is the temperature of the nuclear subsystem. Here we have taken into account the fact that

$$(\mu H_\Delta)^2=\frac{2I(I+1)}{3}\frac{J_0\omega_n^2}{T},$$

and have neglected the frequency dispersion of the relaxation time,[10] in accordance with the smallness of the quantity $(\mu H_\Delta/\varepsilon)^2$. In the region of relatively small wave vectors (i.e., for $k<\varepsilon_0/s$), besides the NSW–NSW scattering processes with a relaxation rate that depends practically linearly on the temperature [see (17)], the above-considered linear NSW → phonon conversion processes also contribute to the NSW relaxation:

$$\Gamma_{n-ph}=\frac{I(I+1)}{6\pi}\beta\frac{\Theta^2}{Mv^2}\left(\frac{\omega_n}{\varepsilon_0}\right)^4\left(\frac{\omega_n}{\mu H_\Delta}\right)^2\left(\frac{J_0}{\omega_D}\right)^3, \tag{15}$$

where

$$\beta=\langle|\varphi'(\mathbf{e},\mathbf{n})|^2\rangle,\quad \omega_D=v/V_0^{1/3}.$$

In contrast to the formula (14), the processes in which the phonons participate make to the NSW relaxation a contribution that depends essentially on the strength of the static magnetic field.

In deriving the expressions (14) and (15) we took into consideration the fact that the nuclear-spin fluctuations are uncorrelated, i.e., that

$$\langle\delta I_{g,f}^z(\mathbf{k}-\mathbf{q})\delta I_{g,f}^z(\mathbf{k}-\mathbf{q})\rangle\approx I(I+1)/3.$$

The expressions (14) and (15) [and also (17) below] allow us to estimate the temperature dependence of the nonequilibrium contribution to the relaxation of the uniform precession of the NSW, $\Delta\Gamma$ [see the formula (21) below].

Thus, both mechanisms of NSW excitation are effective means of creating the nonequilibrium state in the nuclear subsystem in the entire frequency range in which the NSW exist. In this case the populations of the nuclear-spin-wave states are given by the expression (8) when the linear-conversion mechanism obtains, and by the expressions (12) and (13) (for $P>P_c$) in the parametric-excitation mechanism.

The kinetic equation for the NSW occupation numbers that results from the NSW–NSW scattering processes has the form

$$\frac{dn_k}{dt}=32\pi V_0^2\iint\frac{d\mathbf{k}_1\,d\mathbf{k}_2}{(2\pi)^6}|\Phi_{4n}|^2[n_kn_{k_1}(n_{k_2}+n_{k'})-n_{k_2}n_{k'}(n_k+n_{k_1})]\delta(\Gamma_n|\omega_{nk}+\omega_{nk_1}-\omega_{nk_2}-\omega_{nk'});$$
$$\mathbf{k}'=\mathbf{k}+\mathbf{k}_1-\mathbf{k}_2,\quad k_i<k^*. \tag{16}$$

Because of the fact that the inequality $\omega_n\ll T$ is valid in the temperature range in question, the collision integral has been written in the classical limit. Varying (16) with respect to n_k, we have for the relaxation rate in these processes the expression

$$\Gamma_{4n}=16\pi V_0^2\iint\frac{d\mathbf{k}_1\,d\mathbf{k}_2}{(2\pi)^6}|\Phi_{4n}|^2[n_{k_1}(n_{k_2}+n_{k'})-n_{k_2}n_{k'}]\delta(\Gamma_n|\omega_{nk}+\omega_{nk_1}-\omega_{nk_2}-\omega_{nk'}). \tag{17}$$

In the equilibrium case, setting the wave-occupation numbers equal to the thermodynamic-equilibrium values,

$$n_{k_i}=n_{k_i}^0=T/\omega_{nk_i},$$

we obtain for the relaxation of the NSW with $k<\varepsilon_0/s$ the expression[8)]

$$\Gamma_{4n}=\frac{I(I+1)}{2^5\cdot3\pi^3}T\left(\frac{\varepsilon_0}{\mu H_\Delta}\right)^4\left(\frac{\omega_n}{\Theta_N}\right)^3\left(\frac{J_0}{\Theta_N}\right)^3\left(a_1+\frac{2}{3}\ln\frac{T^*}{T}\right)$$
$$T^*=\omega_n(\Theta_N/\varepsilon_0)^{2/3},\quad a_1\sim1 \tag{18}$$

Let us emphasize that the inequality $T\ll T^*$ is equivalent to the condition for the existence of well-defined NSW in the nuclear subsystem.

In the case of an essentially nonequilibrium nuclear subsystem, we obtain from (17) for the additional relaxation $\Delta\Gamma$ of the uniform precession of the nuclear spins the expressions

$$\Delta\Gamma_1=\frac{sk}{2^6\pi}\left(\frac{\omega_n}{\mu H_\Delta}\right)^2\frac{T}{\Theta_N}\left(\frac{J_0}{\Theta_N}\right)^2 N_n, \quad sk\leqslant\varepsilon_0\sqrt{2}, \tag{19a}$$

$$\Delta\Gamma_2=\frac{\omega_n}{2^5}\frac{\omega_n}{\Gamma}\frac{\omega_n^2J_0^2}{\varepsilon_0^4}\left(1+\frac{\varepsilon_0^2}{\varepsilon_k^2}\right)^2 N_n^2, \quad \Gamma=\Gamma(0)+\Gamma(\mathbf{k}). \tag{19b}$$

The expression (19a) corresponds to the case in which one of the NSW in the four-wave interaction process is a nonequilibrium wave; (19b), to the case of two nonequilibrium NSW. A direct comparison shows that $\Delta\Gamma_2 > \Delta\Gamma_1$ when

$$N_n>\frac{1}{4}\frac{\Gamma}{\omega_n}\frac{T}{\omega_n}\left(\frac{\varepsilon_0}{\Theta_N}\right)^3\left(\frac{\varepsilon_0}{\mu H_\Delta}\right)^2\sim 10^{-7}. \tag{20}$$

For relatively high sound-pulse powers we find from the formula (19b) with allowance for (6), (8), and (9) that

$$\Delta\Gamma=\frac{J_0^2}{16\pi^2}\frac{\langle|\hat{U}_{ph-n}|^2\rangle^2}{\Gamma_n^3(\mathbf{k})}\left(\frac{\omega_n}{\varepsilon_0}\right)^4\left(1+\frac{\varepsilon_0^2}{\varepsilon_k^2}\right)^2\left(k^2V_0\Big/\frac{\partial\omega_{nk}}{\partial k}\right)^2 N_{ph}^2. \tag{21}$$

Similarly, using the expressions (12) and (13), we easily find the final expression for $\Delta\Gamma$ in the case of the parametric mechanism of NSW excitation.

The relation (21) contains, besides the quadratic dependence on the number of nonequilibrium phonons, features in respect of the frequency dependence of the nonequilibrium relaxation $\Delta\Gamma$ in two regions of the spectrum: It has a maximum, due to the dependence of $\Delta\Gamma$ on the density of NSW states

$$k^2(\omega)\Big/\frac{\partial\omega_n(\mathbf{k})}{\partial k},$$

in the frequency region $\Omega\approx\omega_n$ and a maximum, due to a magnetoacoustic resonance of the NSW and phonons, in the region $\Omega\approx\omega_{n0}$. The latter follows from the expression (A. III. 2) with allowance for the fact that the coefficient, $u_{n\to ph}$, of the canonical transformation of a NSW into a quasiphonon decreases rapidly as the distance between sound frequency and the frequency corresponding to the intersection of the spectra increases.

If the temperature dependence of the quantities is measured at a constant DFS, then from the relations (1), (14), and (18), namely,

$$\frac{\partial\omega_n(k)}{\partial k}\sim\frac{(\mu H_\Delta)^2}{\varepsilon^4}=\left(\frac{\mu H_\Delta}{\varepsilon}\right)^4\frac{1}{(\mu H_\Delta)^2}\sim T, \quad \Gamma_n\sim T,$$

it follows that $\Delta\Gamma\sim T^3$. This relation corresponds to the experimental conditions of the present work, and agrees, in respect of the temperature dependence, with the measurement results.

Let us give an estimate for the magnitude of the additional relaxation $\Delta\Gamma$ due to the nonequilibrium character of the phonon subsystem of the antiferromagnet. Taking the characteristic parameter values $J_0\sim 30$ K, $Mv^2\sim 10^5$ K, $\varepsilon_0\sim 1$ K, $k\sim 10^5$ cm^{-1}, and $T\sim 1$ K ($\Gamma_n\sim 10^3$ Hz), and assuming for the magnetoelastic energy $\Theta\approx b_1V_0$ (in the notation of Ref. 24) the value 1.6×10^{-1} K, which corresponds to $b_1=2.7\times10^5$ and $V_0=\mu S/M_0=8.3\times10^{-23}$ cm^3, we can write the expression (21) in the form $\Delta\Gamma\sim 10^{11}N_{ph}^2$ Hz. The theoretical $\Delta\Gamma$ values coincide with the experimentally observed values for a nonequilibrium-phonon number of $N_{ph}\sim 3\times10^{-4}$, which, for a pulse power of 1 W and duration $\tau\sim 10^{-6}$ sec, which are characteristic of the experimental conditions of the present work, corresponds to an electromagnetic → sound energy conversion ratio of ~10^{-2}.

6. CONCLUSION

Thus, the experimentally observed acceleration of the transverse relaxation of the uniform precession of the nuclear magnetization under conditions of strong excitation of the phonon subsystem is apparently connected with the following process. The nonequilibrium phonons excite nuclear spin waves at an eigenfrequency (see Fig. 1) and these waves in turn participate in the spin-spin relaxation of the uniform precession. We have considered two mechanisms of NSW excitation by sound: the mechanism of linear conversion of a phonon into a NSW in interaction processes involving the fluctuations of the z component of the paramagnetic nuclear spin and the paramagnetic mechanism, due to a nonlinear process of the type $2ph\to 2n$. The first process is a nonthreshold process, whereas the paramagnetic mechanism is realized at sound-pulse energy flux densities higher than a critical value P_c, which is of the order of 1 – 10 W/cm^2 at liquid-helium temperatures. The contribution $\Delta\Gamma$ experimentally observed in broad ranges of sound-pulse powers and durations corroborates the linear conversion mechanism.

The mechanism of linear conversion of phonons into NSW guarantees a contribution, $\Delta\Gamma$, to the relaxation whose dependence on the sound-pulse power and duration ($\Delta\Gamma\sim N_{ph}^2\sim P^2\tau^2$), sound frequency, and temperature agrees with the experimental dependences. The theoretical $\Delta\Gamma$ value computed for the linear-conversion mechanism coincides in order of magnitude with the value observed in experiment.

The proposed linear-conversion mechanism may be the cause of the acoustic-NMR saturation observed in Ref. 25 in the continuous NSW-excitation regime.

The possibilities of the developed experimental method of investigating the magnon–phonon interactions through the study of the acceleration of the relaxation processes are far from being exhausted, and may later on be used to investigate both the phonon and the magnon subsystems in magnetically ordered media.

In conclusion, the authors express their profound gratitude to A. S. Borovik-Romanov for attention to the work and for useful discussions and to V. A. Golenishchev-Kutuzov, M. A. Savchenko, and V. L. Preobrazhenskii for a number of valuable discussions and comments.

APPENDIX 1

The Hamiltonian of the interaction of the long-wave excitations of both the electronic and nuclear subsystems with the paramagnetic states of the nuclear subsystem can be derived in much the same way as is done in Ref. 10. Separating the interaction of the waves with the fluctuations of the z component of the nuclear spin, we obtain

$$\mathscr{H}^{(2)}=\frac{1}{N}\sum_{\mathbf{k}_1\mathbf{k}_2}[\hat{U}^{(1)}_{m-n}(\mathbf{k}_1,\mathbf{k}_2)(c_{m\mathbf{k}_1}-c^+_{m-\mathbf{k}_1})(c_{n-\mathbf{k}_2}-c^+_{n\mathbf{k}_2}) + \hat{U}^{(2)}_{2n}(\mathbf{k}_1,\mathbf{k}_2)(c_{n\mathbf{k}_1}-c^+_{n-\mathbf{k}_1})(c_{n-\mathbf{k}_2}-c^+_{n\mathbf{k}_2})], \quad \text{(A.I.1)}$$

where

$$\hat{U}^{(1)}_{m-n}(\mathbf{k}_1,\mathbf{k}_2)=\frac{\omega_n^2}{4\mu H_\Delta}\frac{J_0}{(\varepsilon_{\mathbf{k}_1}\omega_{n\mathbf{k}_2})^{1/2}}\hat{\mu}^z_{\mathbf{k}_1-\mathbf{k}_2}, \quad \text{(A.I.2a)}$$

$$\hat{U}^{(2)}_{2n}(\mathbf{k}_1,\mathbf{k}_2)=\frac{\omega_n^3}{4\varepsilon_{\mathbf{k}_1}\varepsilon_{\mathbf{k}_2}}\frac{J_0}{(\omega_{n\mathbf{k}_1}\omega_{n\mathbf{k}_2})^{1/2}}\hat{\mu}^z_{\mathbf{k}_1-\mathbf{k}_2}, \quad \text{(A.I.2b)}$$

are the respective operators, and

$$\hat{\mu}^z_{\mathbf{k}_1-\mathbf{k}_2}=\delta\hat{I}_g^z(\mathbf{k}_1-\mathbf{k}_2)+\delta\hat{I}_f^z(\mathbf{k}_1-\mathbf{k}_2),$$

$$\delta\hat{I}^z_{g,f}(\mathbf{k}_1-\mathbf{k}_2)=\int d\mathbf{r}\exp[i(\mathbf{k}_1-\mathbf{k}_2)\mathbf{r}][\hat{I}^z_{g,f}(\mathbf{r})-\langle I\rangle]$$

are the Fourier transforms of the fluctuations of the longitudinal components of the nuclear spins of the two sublattices.

APPENDIX 2

Let us give the derivation of the effective four-wave amplitudes of the interaction of the phonons with the spin waves and the NSW. As an example, we consider a process of the type $2ph\to 2n$, which is of greatest interest to us here. Amplitudes of processes of this type arise when we consider the three-wave processes in second-order perturbation theory, as well as when we consider the four-wave nonlinear interactions of the NSW with each other and with the spin waves along with the linear coupling of the magnons (SW and NSW) with the phonons. The contributions from the various mechanisms to the $2ph\to 2n$ conversion process are diagrammatically depicted in Fig. 8(b).

An analysis shows that the dominant contribution to the amplitude Φ_{2n-2ph} is made by the first two terms of Fig. 8(b), and has the form

$$\Phi_{2n-2ph}=-\frac{J_0}{8}\frac{\omega_n}{\mu H_\Delta}\left(\frac{\omega_n}{\varepsilon_0}\right)^{1/2}u_{n-ph}\left[u_{m-ph}+\frac{\mu H_\Delta}{\varepsilon_0}\left(\frac{\omega_n}{\varepsilon_0}\right)^{1/2}\left(1+\frac{\varepsilon_0^2}{\varepsilon_\mathbf{k}^2}\right)u_{n-ph}\right], \quad \text{(A.II.1)}$$

$$u_{ph-m}\approx 2G_m/\varepsilon_\mathbf{k}, \quad \text{(A.II.2a)}$$

$$u_{ph-n}=\frac{2G_n}{(\Omega_{1\mathbf{k}}^2-\Omega_{2\mathbf{k}}^2)^{1/2}}\left(\frac{\Omega_\mathbf{k}}{\Omega_{1\mathbf{k}}}\right)^{1/2}\frac{\omega_{n\mathbf{k}}}{(\Omega_{1\mathbf{k}}^2-\omega_{n\mathbf{k}}^2)^{1/2}} \quad \text{(A.II.2b)}$$

are the coefficients of linear conversion into phonons of SW and NSW respectively,

$$G_m=i\Theta\left(\frac{\Omega_\mathbf{k}}{Mv^2}\frac{J_0}{\varepsilon_\mathbf{k}}\right)^{1/2}\varphi'(\mathbf{e},\mathbf{n}), \quad \text{(A.II.3a)}$$

$$G_n=i\Theta\left(\frac{\Omega_\mathbf{k}}{Mv^2}\frac{J_0}{\omega_{n\mathbf{k}}}\right)^{1/2}\frac{\omega_n\mu H_\Delta}{\varepsilon_\mathbf{k}^2}\varphi'(\mathbf{e},\mathbf{n}) \quad \text{(A.II.3b)}$$

are the linear SW–phonon and NSW-phonon coupling constants; $\Omega_{1\mathbf{k}}$ and $\Omega_{2\mathbf{k}}$ are the magnetoelastic-oscillation amplitudes of the NSW and the phonons determined by the dispersion equation

$$(\omega^2-\omega_{n\mathbf{k}}^2)(\omega^2-\Omega_\mathbf{k}^2)=4\Omega_\mathbf{k}\omega_{n\mathbf{k}}|G_n|^2,$$

in which the $\omega_{n\mathbf{k}}$ and $\Omega_\mathbf{k}$ are the unperturbed—by the linear coupling G_n—NSW and phonon frequencies (but with allowance for the coupling with the SW).

The formula (A.II.1) gets simplified when the resonance condition $\Omega_\mathbf{k}=\omega_{n\mathbf{k}}$ is fulfilled, as well as at points far from the resonance, where the inequality

$$(\Omega_\mathbf{k}^2-\omega_{n\mathbf{k}}^2)^2\gg 16\Omega_\mathbf{k}\omega_{n\mathbf{k}}|G_n|^2,$$

is valid. In these particular cases the amplitude Φ_{2n-2ph} assumes the form

$$\Phi_{2n-2ph}=\begin{cases}-\dfrac{J_0}{16}\left(\dfrac{\omega_n}{\varepsilon_0}\right)^2\left(1+\dfrac{\varepsilon_0^2}{\varepsilon_\mathbf{k}^2}\right),\\ -\dfrac{\Theta^2}{2Mv^2}\dfrac{\omega_n^2J_0^2}{\varepsilon_0^4}\dfrac{\Omega}{\Omega-\omega_{n0}}\left[1+\dfrac{\Omega}{\Omega-\omega_{n0}}\left(\dfrac{\mu H_\Delta}{\varepsilon_0}\right)^2\left(1+\dfrac{\varepsilon_0^2}{\varepsilon_\mathbf{k}^2}\right)\right]|\varphi'(\mathbf{e},\mathbf{n})|^2.\end{cases}$$

Owing to the fact that the magnitude of the phonon wave vector satisfies the condition $sq\sim\omega_n s/v\ll\varepsilon_0$, we have neglected the corresponding dispersion in the spin-wave spectrum.

Similarly, we can derive the amplitudes of the other phonon–spin wave and phonon–NSW interaction processes.

The functions, figuring in the formulas (5) and (A.II.3), of the polarization, e, and direction, $\mathbf{n}=\mathbf{q}/|\mathbf{q}|$, of propagation of the phonon have the form

$$\varphi(\mathbf{e},\mathbf{n})=e_xn_x-e_yn_y+\tilde{\xi}(e_yn_z+e_zn_y), \quad \text{(A.II.4a)}$$

$$\varphi'(\mathbf{e},\mathbf{n})=e_xn_y+e_yn_x+\tilde{\xi}(e_xn_z+e_zn_x), \quad \text{(A.II.4b)}$$

where $\tilde{\xi}=B_{14}/2B_{12}$.

Let us give for comparison the formula for the critical energy flux in the course of a first-order instability. Since

$$N_c^{\rm I}=\frac{\Gamma_n(\mathbf{k}_1)\Gamma_n(\mathbf{k}_2)}{2|\Psi_{2n-ph}|^2},\quad \mathbf{k}_1+\mathbf{k}_2=\mathbf{q}_0,$$

taking (5) into consideration, we obtain

$$P_c'=\frac{1}{2}C^{(2)}v\left(\frac{\Gamma_n(k)}{\omega_n}\right)^2\frac{\varepsilon_k^8}{\Theta^2J_0^2(\mu H_\Delta)^4}\frac{1}{|\varphi(\mathbf{e},\mathbf{n})|^2}.$$

Taking the ratio of this relation to the similar expression, (11), for the critical flux, P_c, in a second-order process, we obtain

$$\frac{P_c'}{P_c}=\frac{\Gamma_n(k)}{\omega_n}\frac{\varepsilon_\mathbf{k}^8}{\varepsilon_0^4(\mu H_\Delta)^4}\frac{\Omega}{\Omega-\omega_{n0}}\times\left[1+\frac{\Omega}{\Omega-\omega_{n0}}\left(\frac{\mu H_\Delta}{\varepsilon_0}\right)^2\left(1+\frac{\varepsilon_0^2}{\varepsilon_\mathbf{k}^2}\right)\right]\mathrm{tg}^2\frac{2\pi}{3}.$$

APPENDIX III

The effective Hamiltonian describing the linear conversion of phonons into NSW on the fluctuations of the longitudinal component of the paramagnetic nuclear spin can be written in the form

$$\mathscr{H}^{(2)}_{ph-n}=\frac{1}{N}\sum_{\mathbf{k}\mathbf{q}\lambda}[\hat{U}^z_{ph-n}(\mathbf{q}-\mathbf{k})b_{\mathbf{q}\lambda}c_{n\mathbf{k}}^{+}+\text{H. c.}]. \quad \text{(A.III.1)}$$

Contributing to the operator amplitude, $\hat{U}^z_{ph-n}$, figuring in this expression are the SW–NSW interaction processes and the interaction between two NSW and the fluctuations of the z component of the nuclear spin with allowance for the linear coupling of the SW and, consequently, the NSW with the phonons:

$$\hat{U}^z_{ph-n}(\mathbf{q}-\mathbf{k})=\hat{U}^{(1)}_{m-n}(\mathbf{q}-\mathbf{k})u_{m-ph}(\mathbf{q})+\hat{U}^{(2)}_{2n}(\mathbf{q}-\mathbf{k})u_{n-ph}(\mathbf{q}). \quad \text{(A.III.2)}$$

We emphasize that the second term in (A.III.2) is the dominant term in the vicinity of the point of intersection of the phonon and NSW spectra. As we move away from the point of intersection, the contribution of this term rapidly decreases [see (A. II. 2b)]. Far from the magnetoacoustic resonance, the amplitude of the linear conversion process has a significantly simplified explicit form:

$$\hat{U}^{z}_{ph-n}(\mathbf{q}-\mathbf{k})$$

$$=-\frac{i}{2}\Theta\left(\frac{\omega_n}{\varepsilon_0}\right)^2 \frac{J_0}{\mu H_\Delta}\left(\frac{J_0}{Mv^2}\right)^{1/2}\left[1+2\frac{\Omega}{\Omega-\omega_{n0}}\frac{(\mu H_\Delta)^2}{\varepsilon_0\varepsilon_\mathbf{k}}\right]\varphi'(\mathbf{e},\mathbf{n})\hat{\mu}^{z}_{\mathbf{q}-\mathbf{k}}. \quad \text{(A. III. 3)}$$

As $\Omega\to\omega_n$ $(\Omega-\omega_{n0}\approx\frac{1}{2}\omega_n(\mu H_\Delta)^2/\varepsilon_0^2)$ the contributions of the two coupling mechanisms to the amplitude are of the same order of magnitude.

Here, as above, we have neglected the spatial dispersion due to the finite magnitude of the phonon wave vector. Furthermore, on account of the relative smallness of the DFS, we have set Ω, $\omega_{n0}\approx\omega_n$ everywhere except in the factor $\Omega/(\Omega-\omega_{n0})$.

The Hamiltonian of the indirect interaction between the nuclear and elastic subsystems in the region of applicability of the relation (A. III. 3) has been derived by Silverstein,[26] who, however, did not consider the mechanism of linear conversion of sound into NSW during its interaction with the fluctuations of the z component of the paramagnetic nuclear spins.

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[3)] Here and below we use the system of units in which $\hbar=1$ and $k_B=1$.

[4)] The authors thank S. V. Petrov for making the samples available to them.

[5)] For the feasibility of describing the low-frequency mode of the coupled nuclear–electronic vibrations in $CsMnF_3$ within the framework of the two-sublattice model, see Ref. 11.

[6)] A kinetic equation of the type (7) can also be derived, using the diagrammatic technique for spin operators.

[7)] It makes sense to speak of a steady, or, more precisely, a quasi-steady, state since $T_{ph}\gg T_2$ (see Sec. 3).

[8)] In Ref. 10, the contribution from the $k_{1,2}\sim k^*$ region of phase space is omitted in the computation of the relaxation.

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