Nuclear spin wave research

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(Submitted March 6, 1991; revised June 4, 1991)
Usp. Fiz. Nauk **161**, 1–35 (October 1991)

The present state of experimental and theoretical research on nuclear spin waves in weakly anisotropic antiferromagnetics at liquid helium temperatures is described in this review. We consider the basic measurement methods. The basic models which are used to interpret the experimental data are enumerated. Their merits and drawbacks are discussed.

INTRODUCTION

The term "nuclear spin wave" or "nuclear magnon" is used to denote the elementary excitations of one of the spectral branches of the coupled electron and nuclear spin oscillations in magnetically ordered crystals that is situated in the nuclear magnetic resonance frequency range. The most remarkable property of these excitations is that, at liquid helium temperatures, they are the coupled oscillations of two subsystems which are completely different in their magnetic properties. The electron spins are ordered by an exchange interaction; the equilibrium degree of magnetization of the sublattices has reached a maximum value. On the other hand, the state of the spins of the nuclear subsystem is paramagnetic; the polarization is no more than several percent. As a result of the combined oscillations of these two subsystems, the frequency of the electron magnons increases, and the nuclear magnetic resonance frequency (ω_{n0}) decreases and becomes noticeably lower than the frequency (ω_n) which corresponds to the ordinary Larmor precession of nuclear spins. In other words, there arises the so-called dynamic frequency shift of nuclear magnetic resonance pulling. Thus, an entire band of mixed nuclear and electronic states of nuclear spin waves (ω_{nk}) of width $\omega_n - \omega_{n0}$ appears, and moreover, $\omega_{nk} = \omega_{n0}$ for $k = 0$ and $\omega_{nk} \to \omega_n$ for $k \to \pi/a$ (a is the characteristic size of an elementary cell). These states have a noticeable spatial dispersion in the long wavelength part of the spectrum.

At present, nuclear spin waves have been investigated in the weakly anisotropic antiferromagnetics $MnCO_3$, $CsMnF_3$, $CsMnCl_3$, and $RbMnF_3$. The ^{55}Mn nucleus (spin $I = 5/2$) has a large hyperfine interaction constant A ($\omega_n \propto A$, $\omega_n/2\pi \approx 600$ MHz), and 100% abundance of the magnetic isotope; therefore, compounds with manganese are the most convenient ones for studying nuclear spin waves.

The review is structured according to the following plan. An introduction to the fundamentals of the physics of nuclear spin waves is given in Section 1. The history of the origin of the concept of nuclear spin waves and the development of research in this field up to the 1980s are discussed in Section 2. References to the fundamental publications are also given there. This time interval (≈ 20 years) is picked out as the important period for the accumulation of information about nuclear spin waves. The different methods for investigating nuclear spin waves and the merits and drawbacks of these methods are discussed in Section 3. Section 4 is devoted to the modulation method developed by the authors for investigating nuclear spin waves. The results of nuclear spin wave research obtained in recent years are discussed in Sections 5 and 6, as also are certain problems which arose in the process of investigating nuclear spin waves.

1. THE FUNDAMENTALS OF NUCLEAR SPIN WAVE PHYSICS

The concept of the spin wave, or of its quantum, the magnon, serves as one of the fundamental concepts of the physics of magnetic phenomena. Small oscillations of the magnetic moments of atoms are described by electron spin waves. Electron spin waves are propagated through all of a magnetically ordered crystal by means of an exchange interaction. In the simplest magnetic object, a ferromagnetic substance, there is one branch of the electron spin wave, and in an antiferromagnetic with two sublattices, there are two branches of the electron spin wave. They correspond to the two types of spin deviations for sublattices embedded in each other; to in-phase and anti-phase oscillations. In a weakly anisotropic antiferromagnetic, as a rule, the branch of the in-phase (or "quasi-ferromagnetic" f) electron spin waves is low frequency, whereas the anti-phase ("quasi-antiferromagnetic" a[1]) electron spin waves have higher oscillation frequencies.

If the magnetic atoms have nuclei with non-zero spins, then there exists the so-called hyperfine interaction with the energy **ASI** between the electron shell spin **S** and the spin of the nucleus **I**, which is essentially the result of the contact Fermi and dipole-dipole interactions. It is expedient to distinguish two components in the hyperfine interaction: a static part, which describes the interaction of the longitudinal magnetizations of the electron and nuclear spins, and a dynamical part, which describes the interaction of their "circular" components. The static hyperfine interaction creates a strong effective magnetic field at the nucleus, $\sim 10^5$ Oe, which determines the frequency of the free precession of the nuclear spin, the Larmor (or unshifted) nuclear magnetic resonance frequency ω_n. The nuclear spin $\langle I \rangle$ polarized by this field creates a perturbation on the electron shell, which leads to the appearance of a gap in the electron spin wave spectrum:

$$\Delta\omega_{\mathrm{HFI}} \propto H_\Delta \propto \langle I \rangle^{1/2}.$$

In the approximation being considered one can pick out in the behavior of the nuclear spin two characteristic temperature regions:

1) the extremely low temperature region ($T \ll \hbar\omega_n/k_B \sim 0.01$ K), in which polarization reaches its maximum value $\langle I \rangle = I$, and the level of thermal fluctuations is exponentially small, and

2) the high temperature region ($T \gg \hbar\omega_n/k_B$) with the

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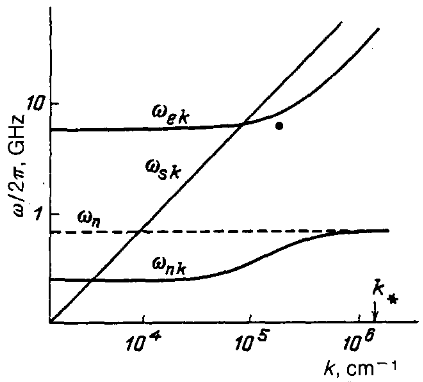

FIG. 1. The Spectra of electron ω_{ek} and of nuclear ω_{nk} spin waves, and of longitudinal sound ω_{sk} in the antiferromagnetic $CsMnF_3$ for $T = 1.7$ K in a field $H = 0.8$ kOe (without allowance for dynamic magnetoelastic interaction).

Curie law for the degree of magnetization $\langle I \rangle \propto h\omega_n / k_B T (\langle I \rangle \ll I)$ and a high level of thermal fluctuations. The upper boundary of this region is determined by the temperature of the transition of the electron spin system from a magnetically ordered state into a paramagnetic state. Here the temperature dependence of the nuclear magnetic resonance frequency indicates the variation of the electron magnetization $\omega_n = A \langle S \rangle / \hbar$.

On neglecting the dynamical part of the hyperfine interaction, the electron spin wave branch ω_{ek} and the free precession of the nuclear spins with the Larmor frequency are the normal oscillations of the magnetic electron-nucleus system. The dynamical part of the hyperfine interaction leads to mixing of these "pure" modes. The branches are "pushed apart" as a result of the linear interaction, and moreover, this separation is larger when the interaction between the pure modes is stronger. Homogeneous ($k = 0$) oscillations are the ones most strongly coupled, and the coupling weakens as the value of k increases. As a result, a branch of mixed nuclear-electron nuclear spin wave oscillations ω_{nk}, which may have a noticeable spatial dispersion, arises in the region of nuclear magnetic resonance frequencies. The electron spin wave spectrum has almost no variation when $\omega_{e0} \gg \omega_n$ (Fig. 1).

In the extremely low temperature region, the width of the nuclear spin wave band $\delta\omega_{n0} \equiv \omega_n - \omega_{n0}$ is determined only by the magnetic field H (for $H \to 0$, $\delta\omega_{n0} \to \omega_n$). In this case, the nuclear spin oscillations are completely correlated and, consequently, nuclear spin waves exist with any k from the Brillouin zone. As the temperature rises, the level of the thermal fluctuations of the nuclear spins gradually increases and the correlation of their motions weakens. Nevertheless, even in a region in which a nuclear subsystem has a small polarization and, accordingly, a high level of fluctuations, there exists a temperature range $h\omega_n / k_B \ll T \ll T_*$ where the long-wavelength nuclear-electron oscillations ($k < k_*$) have a collective nature. The temperature T_* is determined from the condition $\delta\omega_{n0}(T) \sim \Delta\omega_{SN}$, where $\Delta\omega_{SN}$ is the width of an unshifted nuclear magnetic resonance line. At $T \gtrsim T_*$, a nuclear spin wave degenerates into a paramagnetic level, since the dynamic shift of the nuclear magnetic resonance frequency lies within the width of a line. The wave number k_* ($\sim 10^7$ cm^{-1}) is determined from the condition $\delta\omega_{nk} \lesssim 2\gamma_n(k)$, where γ_n is the relaxation rate for a nuclear spin wave. We note that this relation is equivalent to the following condition: the mean free path of a nuclear magnon equals its wavelength. Thus, in the temperature region $\hbar\omega_n / k_B \ll T \ll T_*$, both wave (collective) and also paramagnetic (localized) degrees of freedom appear in a nuclear system.

Antiferromagnetics with anisotropy of the "easy plane" type and with Mn^{2+} magnetic ions are the most convenient objects for the experimental study of nuclear spin waves. Besides the relatively low frequency f-branch of the electron spin waves and the large hyperfine interaction constant for ^{55}Mn, the so-called "exchange amplification" effect appears in these antiferromagnetics (Table I). This effect appears in such a way that the amplitudes of the dynamic interactions (for example, of the hyperfine and magnetoelastic ones) in antiferromagnetics exceed the analogous values in ferromagnetic substances by factors of $(J_0 / h\omega_{ek})^{\nu} \gg 1$ (J_0 is the exchange constant, and $\nu > 0$). In particular, exchange amplification for antiferromagnetics appears as a large dynamic shift of the nuclear magnetic resonance frequency. Furthermore, $T_* \sim (h\omega_n / k_B)(J_0 / h\omega_{e0})^{3/2} \sim 10$ K, as a consequence of which the characteristics of the dynamics of the

TABLE I. The basic parameters of some antiferromagnetics with anisotropy of the "Easy Plane" type with ^{55}Mn.

Parameter	$MnCO_3$	$RbMnF_3$	$CsMnF_3$	$CsMnCl_3$
T_N, K	32.5	82.6	53.5	67
Structure	Rhombohedral	Cubic	Hexagonal	Rhombohedral
Number of magnetic sublattices	2	2	6	18
Density, g/cm^3	3.87	4.68	4.84	3.48
Exchange field H_E, kOe	320	830	350	700
Dzyaloshinskiĭ field H_D, kOe	4.4	0	0	0
Hyperfine interaction parameter, H_Δ^2, kOe^2	$\frac{5.8}{T}$	$\frac{15.6}{T}$	$\frac{6.4}{T}$	$\frac{11.0}{T}$
Inhomogeneous exchange constant α, 10^{-5} kOe·cm	0.78	2.0	0.95	1.3
Volume magnetic atom V_0, 10^{-22} cm^3	0.49	0.70	0.84	1.4
Unshifted NMR frequency, $\omega_n/2\pi$, MHz	640	686	666 667	554 584

nuclear-electron mixed oscillations are observed over a significantly wider temperature interval. In a ferromagnetic substance, the temperature $T_* \sim (\hbar\omega_n/k_B)(J_0/\hbar\omega_{e0})^{3/4} \sim 0.1$ K for iron-yttrium garnet with 100% abundance of the ^{57}Fe isotope.

The nuclear spin wave and electron spin wave spectra in an antiferromagnetic have the following forms:

$$\omega_{nk} = \omega_n[1 - (gH_\Delta/\omega_{ek})^2]^{1/2}, \tag{1}$$

$$\omega_{ek} = g[H(H + H_D) + H_\Delta^2 + (\alpha k)^2]^{1/2}, \tag{2}$$

where $g = 2.8$ GHz/kOe is the magnetomechanical ratio, and H is the external magnetic field present in the basal plane of the crystal.

2. A HISTORICAL OUTLINE OF RESEARCH ON NUCLEAR SPIN WAVES

De Gennes, Pincus, Hartmann-Boutron, and Winter suggested the concept of nuclear spin waves in 1963.[1] By analyzing the oscillations of the electron and nuclear spins in magnetic materials, the authors of Ref. 1 came to the conclusion that, even at temperatures ~ 1 K, a branch of collective spin oscillations with a well-developed dispersion arises near the nuclear magnetic resonance frequency. Microscopically one can describe this phenomenon within the framework of an indirect Suhl–Nakamura interaction,[2,3] which considers a coupling between nuclear spins by means of the exchange of virtual electron magnons. Nuclear spin wave spectra in ferromagnetic and antiferromagnetic substances were calculated in Ref. 1, where it was also predicted that weakly anisotropic antiferromagnetics with ^{55}Mn nuclei, cubic ones ($KMnF_3$) and those with anisotropy of the "easy plane" type ($MnCO_3$), are the most convenient objects for studying nuclear spin waves. In subsequent experimental papers, investigations of nuclear magnetic resonance were carried out on crystals of $CsMnF_3$,[4,5,6] $MnCO_3$,[7] and $KMnF_3$,[8] and the strong dynamic shift of the nuclear magnetic resonance frequency (pulling) in the liquid helium temperature region predicted in Ref. 1 was detected. Here the experimental data were found to be in good agreement with the results of theoretical calculations of the dependence of ω_{n0} on temperature and on the magnitude of the magnetic field (Fig. 2).

Since by means of traditional nuclear magnetic resonance procedure one can carry out investigations only of homogeneous ($k = 0$) nuclear spin oscillations, then to investigate nuclear spin waves with $k \neq 0$, it was natural to consider the possibility of exciting them by a parametric resonance method. However, according to the estimates obtained in Ref. 1, the threshold amplitudes necessary to excite pairs of nuclear spin waves ($\omega_{nk} + \omega_{n-k}$) by a homogeneous microwave field appeared to be experimentally unattainable. The authors of Ref. 1 came to this conclusion by assuming that damping of the nuclear spin oscillations occurs fairly rapidly at a rate $T_2^{-1} \sim \Delta\omega_{SN}$. Ninio and Keffer[9] suggested exciting nuclear spin waves by a combination parametric resonance method in which a microwave field quantum simultaneously generates an electron spin wave + nuclear spin wave pair (the so-called en-process). Their calculation showed that, in this case, due to the direct coupling of the spin oscillations with the alternating field through the electron system, the excitation threshold is significantly reduced and, from estimates for $RbMnF_3$, amounts to $h_c \approx 20$ Oe to 30 Oe, i.e., a value that is experimentally attainable.

This theory stimulated the experiment of Hinderks and Richards,[10] in which they succeeded in parametrically exciting the en-process in $RbMnF_3$. Here the minimum threshold was $h_c \sim 1$ Oe, i.e., the threshold power turned out to be 10^3 times lower than the calculated value. The main result of this paper is the first experimental confirmation of the existence of nuclear spin waves with $k = 0$-10^5 cm^{-1}. The large discrepancy between the measured and calculated values for the threshold indicated that the relaxation rate for nuclear spin waves is lower by at least two orders than the estimate ($\Delta\omega_{SN}/2\pi \sim 1$ MHz) obtained using the second Suhl-Nakamura moment. This fact initially caused amazement, since it seemed that the properties of the paramagnetic nuclear spin subsystem should be completely described within the Van Vleck moment method.

Richards[11] attempted to sort out this situation. In his calculation, he used a relaxation function method, a distinctive modification of the method of moments, but which is applied to the wave degrees of freedom. As a result, analytical expressions were obtained for the rates of relaxation of nuclear spin waves upon their being scattered by the fluctuations of the nuclear magnetization and by electron spin waves. It turned out that the contribution of the second process to the rate of relaxation of a nuclear spin wave is negligibly small by comparison with the first one. One can represent the expression for the rate of relaxation of nuclear spin waves that is determined by the first process (the process of the elastic scattering of nuclear spin waves by fluctuations of

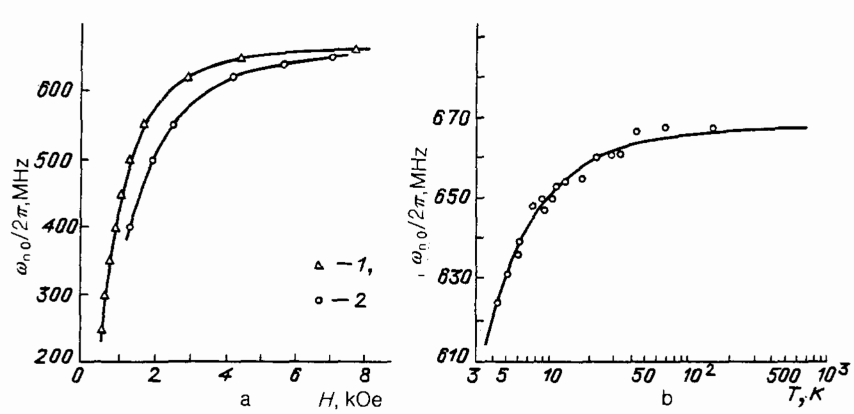

FIG. 2. Dependences of NMR frequency in $CsMnF_3$ on a) the magnetic field at the temperatures 1) 4.2 K and 2) 1.99 K,[5] and b) on temperature.[4]

the longitudinal magnetization of the nuclear subsystem) in the following form:

$$\gamma_R = A_R T k,\ A_R \equiv \frac{\omega_n}{8\pi}\frac{J_0}{k_B\theta_N}\frac{V_0^{1/3}}{\theta_N},\tag{3}$$

where $V_0 \equiv V/\mathcal{N}$, V is the volume of the sample, $\mathcal{N}$ is the total number of magnetic atoms, and $\theta_N \equiv 2\mu_B \propto /V_0^{1/3}k_B$. This equation was obtained using the approximation $\omega_n - \omega_{nk} \ll \omega_n$. Subsequently, the results of Seavey's experiments[12] on the excitation of ee- and en-processes in $CsMnF_3$ confirmed the conclusions of Hinderks and Richards[10] about the low value of the rate of relaxation for nuclear spin waves by comparison with its estimate using the second moment. Furthermore, γ_n turned out to be close in order of magnitude to the estimate by Richards.

By performing a detailed analysis of the expressions for the thresholds of the ee-, en-, and nn-processes, Hinderks and Richards showed[13] that the threshold for the nn-process is easily accessible experimentally.[2)] Such an experiment was first conducted by Adams, Hinderks, and Richards in 1970 on a $CsMnF_3$ sample.[16] The data concerning the relaxation of nuclear spin waves obtained in this, and subsequently in one more experimental paper,[17] were similar in value to the estimates by Richards,[11] but agreement with this theory was not observed for the functional dependences γ_n(T,k) (Fig. 3). Being impressed by the theoretical paper of Woolsey and White,[18] in which a mechanism was suggested for the relaxation of electron spin waves by the inhomogenieties of an antiferromagnetic crystal, the authors of Ref. 17 formulated a hypothesis that inhomogeneities also have a significant effect on the relaxation of nuclear spin waves.[3)]

Almost simultaneously with the experiments on the excitation of a pair of nuclear spin waves by a homogeneous microwave field, Platzker and Morgenthaler[20] considered the possibility of excitation of nn-pairs by hypersound at two frequencies. They performed such an experiment on a $RbMnF_3$ sample at $T = 4.2$ K.[21] However, the experimental dependences $\gamma_n(k,\omega_{nk})$ obtained also departed noticeably from the prediction of the theory of Richards, and therefore here the authors explained the departures by the effect of inhomogeneities of the crystal.

The paper by Weber and Seavey,[22] in which the width of a nuclear magnetic resonance line (i.e., of a nuclear spin wave with $k = 0$) was investigated in $CsMnF_3$ and $RbMnF_3$, served as one more possible argument in favor of the hypothesis of a strong dependence of the damping of nuclear spin waves on crystal inhomogenieties. It was shown in it that the $\gamma_n(H)$ dependence for noticeable dynamic shifts of nuclear magnetic resonance frequency can be described by broadening of a nuclear magnetic resonance line determined by a non-uniformly broadened antiferromagnetic resonance line.

Thus, the first stage of nuclear spin wave research was completed towards the start of the 1970s. Among its main results are:

—the calculation of the nuclear spin wave spectrum and its experimental confirmation for modes with $k = 0$;

—the parametric excitation of nuclear spin waves with $k = 0\text{-}10^6$ cm^{-1} by a microwave field and hypersound, and also the determination of their characteristic lifetimes; and

—the calculation of some mechanisms for nuclear spin wave relaxation.

The insufficiency of describing experimental data by nuclear spin wave relaxation in $CsMnF_3$ and $RbMnF_3$ crystals within the simple theory of Richards led to the hypothesis of the strong effect of crystal inhomogeneities on nuclear spin wave relaxation. As a result, interest in investigating nuclear spin wave relaxation declined for a certain time.

The following important result on nuclear spin wave relaxation was obtained in 1974 in a paper by Yakubovskiĭ[23] devoted to investigating the temperature dependence in $MnCO_3$ of the relaxation rate for nuclear spin waves parametrically excited by parallel pumping at the frequency $\omega_p/2\pi = 875$ MHz. It turned out that, at the temperature $T \approx 3$ K, the linear dependence $\gamma_n \propto T$ changed to the stronger one $\gamma_n \propto T^5$ (Fig. 4). This indicated the presence of at least two processes of relaxation with the rates $\gamma_1 \propto T$ and $\gamma_2 \propto T^5$ $(\gamma_n = \gamma_1 + \gamma_2)$. Analogous temperature dependences were found later by Bun'kov and Dumesh[24] in experiments on the nuclear spin echo in the antiferromagnetics with anisotropy of the "easy plane" type $MnCO_3$ and $CsMnF_3$.

Govorkov and Tulin[25] succeeded in measuring (due to an increase of the pumping frequency to 1200 MHz) the $\gamma_n(k)$ dependence in the range up to $k \approx 10^6$ cm^{-1}. The lin-

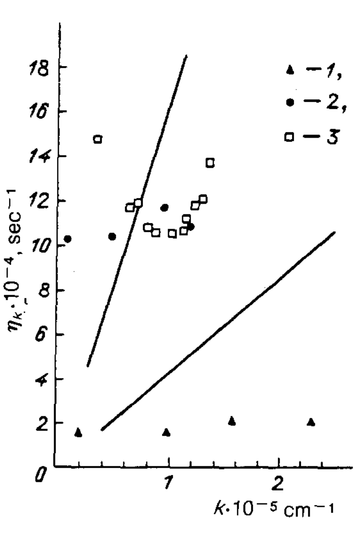

FIG. 3. Dependence of relaxation rate $\eta_k \equiv \gamma_n = 2\pi\Gamma_n$ on wave vector magnitude.[17] *1*) $CsMnF_3$, $T = 1.2$ K; *2*) $CsMnF_3$, $T = 4.2$ K; *3*) $RbMnF_3$, $T = 1.15$ K (values of $0.1\eta_k$ are shown for $RbMnF_3$). The straight lines correspond to the theory of Richards[11] for $CsMnF_3$.

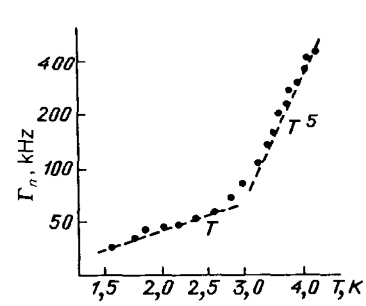

FIG. 4. The temperature dependence of the nuclear spin wave relaxation rate Γ_n with $k = 10^5$ cm^{-1} in $MnCO_3$ for $\omega_p/2\pi \approx 900$ MHz.[23]

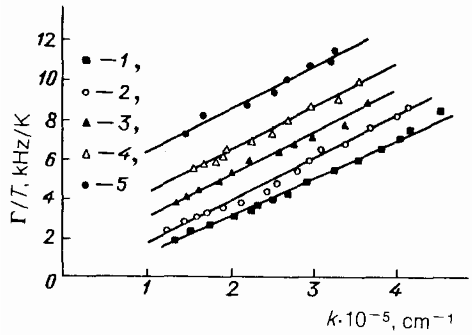

FIG. 5. Experimental results[25] which demonstrate the linear dependence of the nuclear spin wave relaxation rate in $MnCO_3$ on k for $\omega_p/2\pi \approx 1170$ MHz and different temperatures *1*) $T = 1.24$ K; *2*) $T = 1.65$ K; *3*) $T = 3.48$ K; *4*) $T = 3.8$ K; and *5*) $T = 4.14$ K.

ear dependence $\gamma_1 \propto Tk$ predicted by Richards was detected for the first time (Fig. 5). Since the value of γ_1 agreed well with the theoretical prediction, the question about the nature of the process with $\gamma_1 \propto Tk$ was assumed to be solved, but the nature of the process with $\gamma_2 \propto T^5$ remained unclear as before.

A linear dependence of nuclear spin wave damping on temperature and on the magnitude of the wave vector was also detected in a $RbMnF_3$ crystal.[26] However an interpretation of the experimental results on nuclear spin wave relaxation in this antiferromagnetic is difficult, since (in it, unlike $MnCO_3$, $CsMnF_3$, and $CsMnCl_3$) the a-branch of the electron spin waves is situated close to the f-branch, which complicates the dynamics of the nuclear and electron oscillations. Therefore, we do not discuss further nuclear spin wave relaxation in $RbMnF_3$.

The body of knowledge about nuclear spin wave relaxation accumulated up to 1979 was systematized in the review by Tulin.[27] At the same time as this review (mainly of experimental papers), Lutovinov and Safonov published a detailed theoretical paper[28] devoted to nuclear spin wave relaxation. In this paper a separation was suggested of the phase space for the nuclear spin states into two characteristic regions:

1) a region of strictly nuclear spin waves ($k < k_*$) with a well-developed dispersion of ω_{nk}; and

2) a region of localized nuclear spin oscillations ($k > k_*$), in which one can neglect the dispersion of ω_{nk}.[4)] Within such a physically understandable separation, the nuclear magnons were considered as bosons, for calculating which a standard diagram technique was used. As a result of analyzing different intrinsic nuclear spin wave relaxation processes which arise in a model of a two-sublattice antiferromagnetic, the authors of Ref. 28 came to the conclusion that one can explain the experimental law $\gamma_2 \propto T^5$ by a process of merging a nuclear magnon and a phonon into a phonon ($n + ph \rightarrow ph$):

$$\frac{\gamma_{n2ph}}{2\pi} = C_{n2ph} \frac{(1-\xi^2)^2}{g} \left(\frac{k_B}{\hbar}\frac{v_m}{v_s}\right)^2 \left(\frac{k_B\tilde{\Theta}}{Mv_s^2}\right)^2 \frac{T^5}{\theta_N^3 \alpha k}, \tag{4}$$

$$C_{n2ph} = \frac{\pi^2}{20I(I+1)}, \quad \xi = \frac{\omega_{nk}}{\omega_n}, \quad v_m = \frac{2\mu_B}{\hbar}\alpha, \quad k \geq \frac{\omega_n}{v_s};$$

here v_s is the speed of sound, $\tilde{\Theta}$ is a parameter which represents a combination of second order magnetoelastic constants and characterizes the efficiency of magnon-phonon processes in a crystal, $M = \rho V_0$, and ρ is the density of the crystal.

The equation for the Richards process was generalized for arbitrary ω_{nk} in Ref. 28, and a strong frequency dependence was obtained:

$$\tilde{\gamma}_R = \xi^3 \gamma_R. \tag{5}$$

Notwithstanding a certain "heuristic value" of the method used in Ref. 28, rigorous calculations of nuclear spin wave relaxation carried out subsequently[30,31,32] by using the diagram technique for the spin operators confirmed the main conclusions of Ref. 28.

Thus, the second stage of nuclear spin wave research was completed towards the start of the 1980s. The presence in $MnCO_3$ and $CsMnF_3$ of at least two relaxation mechanisms with $\gamma_1 \propto Tk$ and $\gamma_2 \propto T^5$ was shown experimentally. Here γ_1 was attributed to the Richards process, and γ_2 to a process of the interaction of a nuclear magnon with two phonons.

3. METHODS FOR INVESTIGATING NUCLEAR SPIN WAVES

At present, a number of methods is known which may be used to study both the spectrum of nuclear spin waves and also the lifetime of these excitations, which characterizes the efficiency of their interaction with each other, with other quasiparticles, and with crystal defects.

Let us consider the possibilities for investigating the spectrum of nuclear spin waves. As was noted, nuclear spin waves are the coupled oscillations of the electron and nuclear spins, and furthermore, the spatial dispersion of nuclear spin waves is completely determined by the electron component, i.e., the spectrum of nuclear spin waves is, in a definite sense, a "reflection" of the electron spin wave spectrum. This circumstance enables one to form an opinion about nuclear spin waves from the results of electron spin wave research. In accordance with Expression (1), for this it suffices to know the "unshifted" nuclear magnetic resonance frequency ω_n and the parameter H_Δ which characterizes the efficiency of the static hyperfine interaction. One can measure the frequency ω_n by the methods of ordinary nuclear magnetic resonance, of nuclear spin echo, or of double electron-nuclear resonance at the limit of strong H fields and high temperatures T,[4,5] and one can determine the parameter H_Δ from the results of investigating the dependences of the frequencies of antiferromagnetic resonance ω_{e0} or of nuclear magnetic resonance ω_{n0} on H and T.[7]

The electron spin wave spectrum of ω_{ek} is investigated well in the $k \gtrsim 10^6$ cm^{-1} value range by the method of inelastic neutron scattering,[33] and at smaller k values, one can obtain information about the electron spin wave spectrum by means of optical methods.[34]

An interesting possibility for studying the electron spin wave spectrum exists in antiferromagnetic crystals in which the speed of sound v_s exceeds the "limiting" electron spin wave speed v_m (almost all the crystals investigated by us are among these). In this case there exists a point of intersection for the spectra of sound and of the electron spin waves with $k = 0$, the position of which depends on H and T (see Fig. 1). In a paper by Seavey[35] devoted to the investigation of parametrically excited electron spin waves in $CsMnF_3$, peaks were detected in the dependence of the electron spin wave relaxation rate on k which correspond to the points of intersection of the electron spin wave spectrum with the branches of longitudinal and transverse sounds. If one knows the frequency of the excited electron spin waves and the speeds of these sounds, one can easily determine the value of k for the intersection point. Later on, Kotyuzhanskiĭ and Prozorova[36] successfully used this method over a broad frequency range.

The set of methods enumerated enabled one to investigate the electron spin wave spectrum sufficiently well in a number of antiferromagnetics and to confirm its agreement with Expression (2), which also was an indirect confirmation of the validity of Eq. (1).

Nevertheless, a direct experimental verification of the nuclear spin wave spectrum seems interesting. The difficulty of this task is caused by the fact that not one of the enumerated methods of investigating an electron spin wave spectrum with $k \neq 0$ is practically suitable for nuclear spin waves, since their spectrum is concentrated in a comparatively low frequency region ($\omega_{nk}/2\pi \lesssim 10^9$ Hz), which differs greatly from the frequencies of visible light ($\sim 10^{15}$ Hz), and the wave numbers $k < 10^6$ cm^{-1} are significantly lower than for thermal neutrons ($\sim 10^8$ cm^{-1}). Ozhogin and Safonov suggested[37] a method of investigating a nuclear spin wave spectrum by the inelastic scattering of hypersound with frequencies from 1 GHz to 10 GHz; however, such experiments have not yet been performed.

The study of the nuclear spin wave dispersion law by the spectrum intersection point is also impossible, since the intersection of a nuclear spin wave and sound occurs at the too low $k \lesssim 10^4$ cm^{-1} values (see Fig. 1). Nevertheless, we think that we succeeded in $CsMnF_3$ in obtaining direct experimental confirmation of Expression (1) for a nuclear spin wave spectrum in the $k \sim 10^5$ cm^{-1} range of values.[38] This will be considered in more detail in Section 5, but for now let us note only that the experimental verification of the nuclear spin wave spectrum became possible due to the noticeable effect of the width of the electron spin wave spectrum on the damping of nuclear spin waves. As a result, the phonon peaks in the electron spin wave relaxation rate which correspond to the intersection of electron spin wave spectra and sound also appear in nuclear spin wave relaxation with the same k values.

One more method of verifying a nuclear spin wave spectrum, which is apparently simple to carry out experimentally, is of interest. Two microwave fields, one of which excites a combination en-process, and the second serves as a probe to determine the frequency of the nuclear spin waves that are excited, are supplied to the sample. Here the magnitude of the wave vector $|k_{\rm NSW}| = |k_{\rm ESW}|$ is calculated from the known spectrum of electron magnons.

Let us now discuss methods for studying the nuclear spin wave relaxation rate. Generally speaking, the width of an ordinary nuclear magnetic resonance line must give information about the relaxation rate of the uniform precession of the nuclear magnetization ($k = 0$). However, a noticeable non-uniform broadening of a nuclear magnetic resonance line[39] caused by defects in the sample (including irregularities of its surface) does not allow one to extract information from the width of a nuclear magnetic resonance line about the intrinsic (i.e., inherent to an ideal crystal) processes of nuclear spin wave relaxation with $k = 0$.

The nuclear spin echo method,[24,40,41] which, upon the fulfillment of definite requirements for the power and duration of microwave pulses and also for homogeneity of the microwave field in the volume of the sample, enables one to measure the times of spin-spin (T_2) and spin-lattice (T_1) relaxation of the nuclear magnetization precession, is significantly more informative. However an extensive packet of nuclear spin waves with $k = 0$-10^4 cm^{-1} is excited during the use of this procedure, and the relaxation times measured represent an integral characteristic $\gamma_n = (2T_2)^{-1}$ of this packet, which hinders the physical interpretation of the results obtained.

At present, a method of parametric excitation of these quasiparticles by a microwave pumping field $h \cos\omega_p t$ parallel to a constant magnetic field H is the most efficient method for investigating nuclear spin wave relaxation. The parallel pumping method was suggested in 1961 and was achieved at the same time in ferromagnetic iron-yttrium garnet.[42,43] Later on, it earned a favorable reputation for investigating electron spin waves in ferrites and antiferromagnetics. Its main merits are the excitation of a narrow wave packet ($\Delta k \ll k$) and the possibility to control the value of k within broad limits by means of varying H ($2\omega_k(H) = \omega_p$). A drawback of the method is the need for absolute measurements of the threshold amplitude of the magnetic microwave field h_c. Just this quantity is connected with the relaxation rate γ of the quasiparticles that are excited.

Methods exist for investigating the relaxation of paramagnetic nuclear spin waves that are not connected with measurement of the threshold h_c. Observation of the drop of the induction signal from a system of paramagnetic nuclear spin waves after a rapid shutoff of a microwave pumping pulse is the most direct method.[39] However, one observed it only at very low temperatures ($T \leqslant 1.2$ K) in a region of small k. Actually the author of Ref. 39 used this method only to check the results of calculating the nuclear spin wave relaxation rate for the threshold h_c.

Another method of measuring nuclear spin wave relaxation (the spin echo for paramagnetic nuclear spin waves) was suggested and achieved by Govorkov and Tulin.[44] The results they obtained for nuclear spin wave relaxation in $MnCO_3$ agree well with the values calculated for the threshold h_c.[45]

The magnetic field modulation method opens interesting possibilities for measuring the nuclear spin wave relaxation rate. Studying the effect of modulation on the threshold h_c and investigating the frequency dependence of the modulation response of a nuclear spin wave system to a modulation field[46,47] enable one to measure the nuclear spin wave relaxation rate fairly accurately.

All these methods (technically and with respect to data reduction methods) are significantly more complicated than the simple, rapid, and reliable method of measuring the threshold h_c for the parametric process. Therefore, in investigating linear relaxation, we used them only to calibrate the measurement results for nuclear spin wave relaxation with respect to the threshold h_c.

In concluding this section, we recall two more methods. Besides the parametric process of exciting a nuclear spin wave pair (the nn-process), the excitation of a nuclear spin wave in a pair with an electron spin wave (the en-process) is possible.[10,15] The value of the nuclear spin wave relaxation rate enters into the equation for the en-process threshold in combination with the corresponding characteristics of the electron spin waves that are excited, which complicates the analysis of the data obtained. Therefore, the investigation of "degenerate" (ee- and nn-) processes turned out to be significantly more convenient for measuring the relaxation of electron and nuclear magnons.

Besides nuclear spin wave pumping with a homogeneous microwave field, the possibility exists for linear and nonlinear excitation of nuclear spin waves with hypersound of suitable frequencies.[20] In principle, the linear excitation of nuclear spin waves with hypersound enables one to investigate the nuclear spin wave spectrum in the region of small $k \sim 10^4$ cm^{-1} and the characteristics of the interaction of sound and nuclear spin waves. And nonlinear excitation is the parametric pumping of a pair of nuclear spin waves with approximately (of the order of k of the sound pumping wave vector) equal frequencies and wave vectors.[21] These methods are very complicated, and it is expedient to use them mainly to study magnetoelastic interactions in those crystals in which the characteristics of nuclear spin waves and phonons had been previously investigated by other methods.

Thus, the method of measuring the h_c threshold with microwave pumping for the parametric process is the simplest and most convenient method for studying nuclear spin wave relaxation. The excitation of nuclear spin waves with well known values of frequency and wave number k enables one to investigate easily the dependence of γ_n on these parameters and on temperature.

Since the calculation of the nuclear spin wave relaxation rate from experimental data is connected with the use of a definite equation, we think it necessary to discuss the question of the validity of using it. A calculation based on a consideration of energy balance gives the following equation for calculating the γ_n of parametric nuclear spin waves:

$$\gamma_n = h_c |V_k| = \gamma_{ei} + \gamma_{nel}, \tag{6}$$

where V_k is the coefficient of nuclear spin wave coupling with the pumping ($V_k = -1/2\partial\omega_{nk}/\partial H$), γ_{el} and γ_{nel} are the relaxation rates for elastic processes (i.e., with no change of the wave energy and with a change of only the direction of wave motion) and for inelastic processes, respectively. In this calculation, the contributions of all processes to the total relaxation of the spin wave under consideration are considered to be additive.

A modification of Eq. (6) based on an assumption of a special role for elastic relaxation processes was suggested in the theoretical Refs. 48 and 49. The result is the same as Eq. (6) in the case when $\gamma_{nel} \gg \gamma_{el}$, however, in the opposite limiting case, a different equation is found:

$$\gamma_n = h_c |V_k| = (\gamma_{el}\gamma_{nel})^{1/2}. \tag{7}$$

The results of analyzing experimental data obtained at sufficiently low temperatures, where elastic processes are dominant for electron spin wave and nuclear spin wave relaxation (their scattering by paramagnetic nuclear spins and the sample boundary) indicate the applicability of Eq. (6). In particular, it has been experimentally demonstrated that, at sufficiently low temperatures ($T \lesssim 2$ K), the main contribution to nuclear spin wave relaxation has the form $\gamma_1 \propto Tk$ in all research on antiferromagnetics with anisotropy of the "easy plane" type ($MnCO_3$, $CsMnF_3$, and $CsMnCl_3$).[25,50,51] As was already noted, this contribution to relaxation is determined by the process of elastic scattering of parametric nuclear spin waves by fluctuations of the longitudinal component of the nuclear magnetization.[11] Reduction of the experimental results using Eq. (6) enables one to obtain good agreement with theory not only for dependences on the basic parameters (T,k), but also for the amount of relaxation for all materials investigated. The use of Eq. (7) does not enable one to obtain the indicated functional dependences. A demonstration of the validity of Eq. (6) has been obtained in the experiments of Govorkov and Tulin by direct measurement of the nuclear spin wave relaxation rate in $MnCO_3$ by a spin echo method for parametric nuclear spin waves.[45]

Thus, the experimental results obtained indicate the additive nature of the contributions of all processes and the validity of using Eq. (6).

4. A MODULATION METHOD FOR RECORDING A THRESHOLD

A block diagram of a decimeter range microwave spectrometer with which an investigation of the parametric excitation of nuclear spin waves was carried out is presented in Fig. 6.

A spiral cavity, which is a half-wave vibrator coiled into a spiral at the ends of which reflection of an electromagnetic wave occurs, was used as a measurement cell. The inner diameter of the spiral equals 8 mm, the coil spacing is 2 mm, and the diameter of the copper conduit is from 0.5 mm to 1 mm. To a first approximation (without allowance for capacitances between winding turns), the conduit length needed to make the spiral is $l = 1/2\lambda$. In such a resonance structure, the magnetic microwave field h is directed along the axis of the spiral. Here the distribution of the fields in the funda-

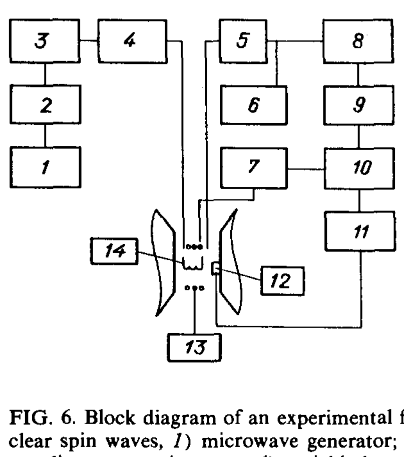

FIG. 6. Block diagram of an experimental facility for investigating nuclear spin waves, *1*) microwave generator; *2*) precision attenuator; *3*) standing wave ratio meter; *4*) variable length line; *5*) rectifier; *6*) spectrum analyzer; *7*) modulation generator; *8*) microwave range receiver; *9*) resonance modulation signal amplifier; *10*) synchronous detector; *11*) two-coordinate automatic recorder; *12*) Hall sensor; *13*) modulation coil; *14*) resonator.

mental oscillation mode is independent of the azimuthal angle, and it varies sinusoidally along the axis and has a fairly uniform distribution over radius (a Bessel function).[39]

The capacitance coupling of the coaxials with the cavity is achieved by stubs placed near its ends. Such coupling is simpler in design than an inductive one, and what is most significant, in this case there is no induction from the modulation coil onto the output coaxial. The input coupling was set close to the critical value ($\beta_{in} \approx 1$), and the output coupling coefficient was $\beta_{out} \ll 1$. The loaded Q-factor of the cavity for the critical input coupling is half its intrinsic Q-factor, and at liquid helium temperatures, it usually was $Q_{ld} = 1/2Q_0 = 300\text{-}500$.

The modulation field at the sample was created by a coil of 2 cm diameter situated coaxially with the spiral cavity. The constant (H), microwave (h), and modulation (H_m) magnetic fields were parallel to each other and were present in the basal plane of the crystal being investigated. The amplitude of the field H_m was varied in a range up to 5 Oe, and the modulation frequency $\omega_m/2\pi$ was varied up to 600 kHz.

Unlike three-dimensional cavities, in which the field distribution is well known for each specific mode and the calculation of the field h for a small sample does not present difficulties in principle, for a spiral cavity (especially without external shielding, as in this case), there is no such definiteness. This is mainly connected with the poorly known field distribution outside the spiral and with the presence of a dielectric housing. Therefore, to calibrate the field h in a spiral cavity, we used the phenomenon of saturation of the electron paramagnetic resonance signal in paramagnetic DPPH. We estimate the magnitude of the absolute error in determining the field h as equal to $\pm 25\%$.

Both pulsed and also continuous procedures are used to determine the threshold amplitude h_c of the magnetic microwave field. In the first case, with a constant H value, one increases the microwave power and records the instant when the characteristic distortion of the shape of the pumping pulse appears. This method is convenient for investigating parametric electron spin waves when the relative change of power $\Delta P/P$ at the output of a cavity with a sample during the excitation of a parametric process is fairly large and amounts to $\sim 10\%$. For nuclear spin waves, this value is approximately 300 times smaller, i.e., 10^{-4} to 10^{-3}, which practically eliminates the possibility for an accurate measurement of h_c in the pulsed regime. This difference is explained by the fact that $\Delta P/P \propto \chi'' h^2 \omega_p Q$, where χ'' is the imaginary part of the nonlinear susceptibility, ω_p is the pumping frequency, and Q is the Q-factor of the measurement cell. The first two parameters are similar for electron spin waves and nuclear spin waves $h_c^{nn} \approx h_c^{ee} \sim 0.1$ Oe, $\chi'' \sim 0.001$), the pumping frequency is by a factor of 20 to 40 lower for nuclear spin waves, and the Q-factors for decimeter range measurement cells (these are usually spiral cavities) are an order of magnitude smaller than the Q-factors for centimeter range three-dimensional cavities. Therefore, one usually uses a continuous procedure to record the threshold h_c for exciting nuclear spin waves (one records the absorption signal during a slow sweep of H for a constant microwave power level).

The values of the field which limit the absorption curve correspond to the threshold of the parametric process for an established value of the microwave field. Such a tracing is shown in Fig. 7a for conditions when sensitivity of recording apparently close to maximum was attained (signal compensation was used at the output of the microwave detector).[25] As is evident from this tracing, even such sensitivity does not allow one to determine reliably the start of the parametric process, and one has to extrapolate the absorption signal to the zero level, for example, with a straight line, as was done in Ref. 25. However, generally speaking, such an extrapolation is ambiguous, since the absorbing power is not necessarily linear with H. As it turned out, the values of H_c obtained by such an extrapolation turned out to be systematically too high. If such an error in determining H_c does not very greatly distort the nature of the measured dependence $\gamma(H)$ for the $MnCO_3$ sample investigated in Ref. 25 with $H_D = 4.4$ kOe by virtue of the fact that a combination of fields $2H_c + H_D$ enters into the equation for the threshold h_c (and furthermore, $H_c < H_D$), then the degree of distortion of the dependence $\gamma(H)$ measured increases sharply in crystals with $H_D = 0$.

To remove the uncertainty in extrapolating to zero absorption, one must significantly increase the sensitivity of recording the absorption signal beyond the threshold of the paramagnetic process. This turned out to be possible due to the use of a modulation method for recording a threshold

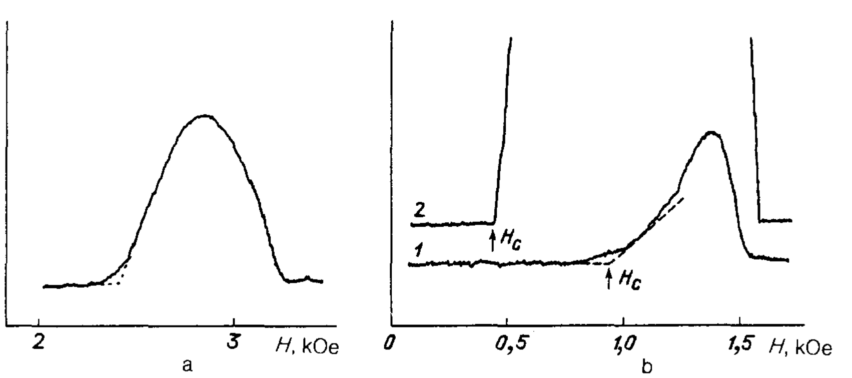

FIG. 7. Tracing of absorbed power a) during microwave pumping of nuclear spin waves in $MnCO_3$ at $\nu_p = 1170$ MHz, $T = 1.24$ K,[25] and experimental tracings of a modulation signal b) during parametric excitation of nuclear spin waves in $CsMnF_3$ (Ref. 47) obtained with the same amplitudes of the microwave pumping field: *1*) the sensitivity is reduced by a factor of approximately 300 (i.e., it approximately corresponds to the possibilities for traditional recording methods), and *2*) the actual sensitivity of the facility.

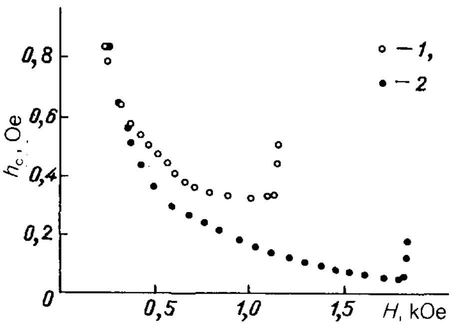

FIG. 8. Dependence of the threshold amplitude h_c for the parametric excitation of nuclear spin waves in $CsMnF_3$ on the magnetic field for $\nu_p = 904$ MHz and the temperatures *1*) 4.2 K and *2*) 1.65 K.

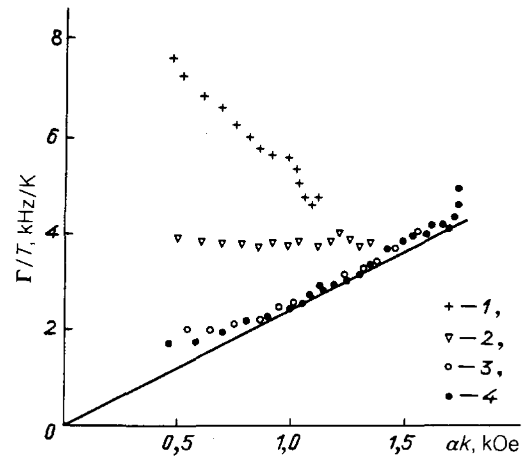

FIG. 9. Dependences of the nuclear spin wave relaxation rate on the wave vector magnitude in $CsMnF_3$ in (Γ/T, ak) coordinates for $\nu_p = 904$ MHz and temperatures *1*) $T = 4.2$ K, *2*) $T = 2.90$ K, *3*) $T = 2.05$ K, and *4*) $T = 1.65$ K.

developed by us,[50,52] the essence of which is reduced to the following. In addition to the constant H and microwave ($h \cos \omega_p t$) fields, one more magnetic field $H_m \cos \omega_m t$ is applied to the sample ($H_m \| H \| h, \omega_m \ll \omega_p$). As was discovered in Ref. 52, signals at the frequencies $\omega_p \pm \omega_m$ appear in the spectrum of the microwave signal at the output of the cavity immediately beyond the threshold of the parametric process, which indicates modulation of the parametric absorption signal by the frequency ω_m. These side bands are easily visible on the screen of the spectrum analyzer. To record precisely the start of the parametric process, the signal from the output of the microwave detector was amplified by resonance at the frequency ω_m and detected synchronously, and after this it was recorded on a two-coordinate automatic recorder as a function of H.

Examples of experimental tracings of this signal are shown in Fig. 7b. The signal/noise ratio for curve 1 corresponds approximately to that attained in Ref. 25, i.e., for a traditional procedure of recording the threshold (in recording this curve, we reduced the sensitivity of the scheme by weakening the useful signal with an attenuator at the input of the detector). As the sensitivity increases (the signal attenuation is decreased), the start of the parametric instability recorded by the scheme is shifted to weaker fields and reaches the value H_c (see Fig. 7b, curve *2*). Since a further increase of sensitivity does not lead to a change of H_c, then it is natural to think that the field H_c is the true start of the parametric excitation of nuclear spin waves at this level of microwave power.[5)] It is evident that the value for the limiting field $\tilde{H}_c$ of the parametric process that is obtained at ordinary sensitivity is almost two times too high by comparison with the value of the "true" H_c.

The magnitude of the absolute error of measuring h_c is fairly large ($\pm 25\%$) for any recording procedure, since it is connected with the errors of measuring the microwave power, the coefficient of coupling with the cavity, and of its Q-factor (an additional error is due to the accuracy of calibrating the field h in the cavity). However the absolute error does not play a special role in comparing the experimental functional dependences for the nuclear spin wave relaxation rate at a fixed pumping frequency with theory, and the relative error that is determined by the stability of the microwave generator level and the accuracy of the attenuator calibration is no more than $\pm 2\%$ in this case.

5. INVESTIGATION OF THE SPECTRUM AND RELAXATION OF NUCLEAR SPIN WAVES

The results of nuclear spin wave research obtained mainly by the authors of this paper from 1982 through 1987 are discussed in this and the following sections.

The experiments reduce to measurement of the dependences of the threshold field h_c on H at different temperatures. Typical $h_c(H)$ dependences at two temperatures are shown in Fig. 8. Such curves give little information by themselves; therefore, from here on we shall analyze the $\Gamma(k)$ dependences calculated from $h_c(H)$. The parameter $\Gamma = \gamma_n/2\pi$ is determined from the threshold Eq. (6), and the value of the wave number k is calculated from the nuclear spin wave spectrum (Eq. (1)) for the condition that $\omega_{nk} = \frac{1}{2}\omega_p$. The dependences of the nuclear spin wave relaxation rates on the magnitude of the wave number in ($\Gamma/T, \propto k$) coordinates in $CsMnF_3$ and $CsMnCl_3$ are shown in Figs. 9 and 10. Such a choice of coordinates enables one to detect any additions to the main relaxation mechanism at low temperatures with $\Gamma_1 \propto Tk$. At temperatures

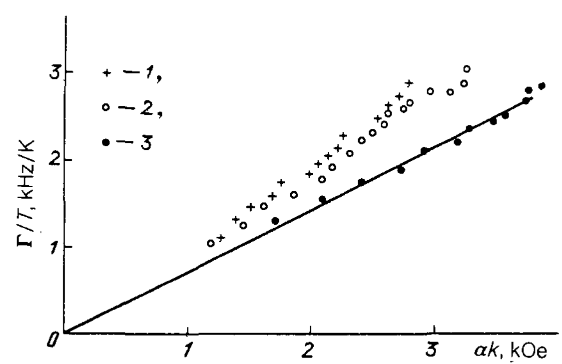

FIG. 10. Dependences of the nuclear spin wave relaxation rate on the wave vector magnitude in $CsMnCl_3$ in (Γ/T, ak) coordinates for $\nu_p = 1040$ MHz and temperatures *1*) $T = 4.2$ K, *2*) $T = 3.1$ K, and *3*) $T = 2.0$ K.

$T \leqslant 2.2$ K, the experimental points in these coordinates lie on one straight line which emerges from the origin of coordinates.

With a further temperature rise, the experimental points start to depart noticeably from this line (see Fig. 9), which indicates an additional contribution to the total nuclear spin wave relaxation in $CsMnF_3$ from at least one more mechanism with significantly different dependences on k and T. By subtracting the corresponding values of Γ_1 from the value of the total relaxation Γ, we obtain the value of the relaxation rate Γ_2 for the additional process. We note that the efficiency of the relaxation process with Γ_2 is low in $CsMnCl_3$ (see Fig. 10).

One must note that, at small values of the wave number ($k < k_{nw}$), the value of Γ becomes independent of k (with an increase of pumping frequency in the 600 MHz to 1200 MHz range, k_{nw} increases from $0.5 \cdot 10^5\ cm^{-1}$ to $10^5\ cm^{-1}$). This is the region of the so-called nonuniform broadening which was observed earlier in $MnCO_3$ [25] and $RbMnF_3$. [39] The experimental results obtained only in the region $k > k_{nw}$ will be considered next.

5.1. The study of features in the nuclear spin wave relaxation rate as a method for checking their spectrum

A feature in the form of a small peak (Fig. 11) whose position $\tilde{k}$ changed with changing temperature T was detected on the smooth dependences of Γ on k and T in the antiferromagnetic $CsMnF_3$. It was noticed that the dependence $\tilde{k}(T)$ is described well by the equation

$$v_s \tilde{k} = \omega_{e\tilde{k}}, \tag{8a}$$

where v_s is the speed of the transverse sound and $\omega_{e\tilde{k}}$ is the frequency of electron spin waves with the wave number $\tilde{k}$ that is calculated from the condition $\omega_{nk}(\omega_{ek}) = \frac{1}{2}\omega_p$, or from $(gH_\Delta/\omega_{ek})^2 = 1 - \xi^2$, which is equivalent to it. Thus, one can represent Relation (8a) in the following form:

$$\alpha \tilde{k} = \frac{H_\Delta}{(1-\xi^2)^{1/2}} \frac{\alpha g}{v_s}, \tag{8b}$$

where $H_\Delta \propto T^{-1/2}$.

It is shown in Fig. 12 that the experimental dependence $\tilde{k}(T)$ (the open circles) fits the line $\tilde{k} \propto H_\Delta$ well. Here the slope of the line corresponds to the ratio $\tilde{k}_{exp}/\tilde{k}_{theo} = 1.0 \pm 0.15$ for the experimental values of the parameters for $CsMnF_3$: $\alpha = (0.95 \pm 0.1) \cdot 10^{-5}$ kOe·cm, [36] and $v_{s\perp} = (2.33 \pm 0.03) \cdot 10^5$ cm/sec. [53] The experimental dependence $\tilde{k}(T)$ for the peak detected in the narrow frequency range $\nu_p = 760$-790 MHz (see Section 6) is shown by the filled circles in Fig. 12. These points also fit the theoretical dependence of Eqs. (8) well, but here the value of the speed of the longitudinal sound fits better as v_s. Thus, for $v_{s\parallel} = (4.6 \pm 0.03) \cdot 10^5$ cm/sec, [53] we find $\tilde{k}_{exp}/\tilde{k}_{theo} = 1.2 \pm 0.2$.

It is obvious that Eq. (8a) is the condition for the intersection of the electron spin wave spectra and the sound. The usual separation of the branches occurs at this point which, however, has no effect on the nuclear spin wave spectrum (no features in ω_{nk} arise at $k = \tilde{k}$). A peak in the electron spin wave relaxation rate γ_{ek} is observed experimentally at the points of intersection of the spectra, [35,36] using which, as was already mentioned in Section 3, the parameters of the electron spin wave spectrum were studied. The natural hypothesis arises that a peak in the nuclear spin wave relaxation is caused by a peak in the electron spin wave relaxation. One can suggest two mechanisms which explain this effect.

The first mechanism arises from the fact that, at the intersection point of the electron spin waves and sound, the electron spin oscillations are strongly mixed with elastic oscillations. The nuclear spin waves, which have a significant part of the electron component within them, thus acquire, in the vicinity of the point k, a stronger coupling with the phonons. However, as a consequence of the large frequency difference $\omega_{nk} \ll \omega_{ek}$, the contribution of the elastic subsystem to the nuclear spin wave relaxation will be attenuated by $(\tilde{\Delta}\omega/\omega_{ek})^2 \ll 1$, where $\tilde{\Delta}\omega$ is the frequency width of the magnon-phonon peak. According to estimates, the contribution of this mechanism for the origin of the peak to nuclear spin wave relaxation is negligibly small.

The second mechanism for the origin of the peak is based on the following idea. Nuclear spin waves are the normal mode of the coupled oscillations of the two subsystems

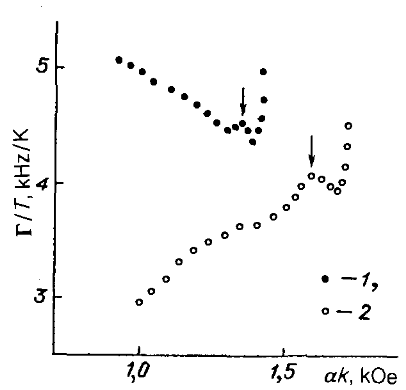

FIG. 11. Behavior of the nuclear spin wave relaxation rate in $CsMnF_3$ in $(\Gamma/T, \alpha k)$ coordinates at the pumping frequency $\nu_p = 1022$ MHz at temperatures of *1*) 4.23 K and *2*) 3.03 K. The positions of "transverse" magnon-phonon peaks are shown by arrows. The rapid increase of the relaxation rate at large k values (i.e., at small H values) is probably caused by the scattering of nuclear spin waves by the domain walls.

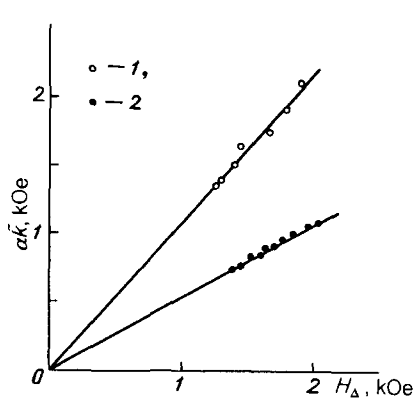

FIG. 12. Temperature dependence of the positions in $(\alpha\tilde{k}, H_\Delta)$ coordinates of *1*) the transverse ($\nu_p = 1022$ MHz) and *2*) the longitudinal ($\nu_p = 775$ MHz) magnon-phonon peaks in nuclear spin wave relaxation in $CsMnF_3$, where the theoretical dependences must have the form of straight lines emerging from the origin of coordinates (see Eq. (8b)).

of electron and nuclear spins, and furthermore, the spatial dispersion in the nuclear spin wave spectrum is completely "transformed" from the electron subsystem. Generally speaking, in addition to the renormalization of the spectra of the initial "pure" modes, their relaxation parameters must also be renormalized. It is obvious that the normal coupled oscillations become damped if even one of the component pure modes has a finite spectral width.

A phenomenological treatment of the idea under discussion based on an analysis of the line shapes of the coupled oscillations was suggested in Ref. 38. A simple equation for the contribution to the nuclear spin wave relaxation introduced from the electron branch was obtained using a weak coupling approximation:[6)]

$$\gamma_n^{(e)} = (d\omega_{nk}/d\omega_{ek})\gamma_e,$$

$$\frac{d\omega_{nk}}{d\omega_{ek}} = \frac{(1-\xi^2)^{3/2}}{\xi}\frac{\omega_n}{gH_\Delta}, \quad \xi \equiv \frac{\omega_{nk}}{\omega_n}. \qquad (9)$$

The value of the increase of the nuclear spin wave relaxation rate at the peak calculated according to Eq. (9) (see Fig. 11), and which has been obtained from the known value of the peak of the electron spin waves[35,36] at the point of their intersection with the transverse sound branch agrees well with the experimental value of 0.5-1 kHz. The fact that no feature corresponding to the intersection point of the electron spin waves with the longitudinal sound branch is observed in Fig. 11 is explained by the fact that its magnitude calculated according to Eq. (9) is approximately an order of magnitude smaller than for the observed "transverse" peak, i.e., it lies within the measurement error for the nuclear spin wave relaxation rate. (We shall consider a discussion of the feature in the nuclear spin wave relaxation that is observed at the pumping frequencies $\nu_p = 760$-790 MHz in Section 6.)

One may consider the results of the investigation of the magnon-phonon peaks in nuclear spin wave relaxation as the first experimental verification of the calculated nuclear spin wave spectrum on the condition that the hypothesis indicated above about the renormalization of the relaxation of coupled oscillations is true.

5.2. The nature of the process of nuclear spin wave relaxation with $\gamma_1 \propto Tk$

We now discuss the results of investigating nuclear spin waves at relatively low temperatures ($T \lesssim 2$ K), when a linear dependence of relaxation rate on temperature and wave number is observed with good accuracy. As was already noted (see Section 2), such behavior of nuclear spin wave relaxation is described well by Eq. (3) for the Richards process, the elastic scattering of nuclear magnons by fluctuations of the longitudinal nuclear magnetization. However the generalization of this equation to the case of arbitrary ω_{nk} (Eq. (5)), which has been carried out in Ref. 28, predicted a strong frequency dependence for this process: $\tilde{\gamma}_R = \xi^3\gamma_R$, $\xi = \omega_{nk}/\omega_n$. After measurements of the $\gamma_1(\omega_{nk})$ dependence in three antiferromagnetics over a fairly broad frequency range (in $CsMnF_3$ for $\xi = 0.45$-0.9,[50] in $CsMnCl_3$ for $\xi = 0.71$-0.93,[51] and in $MnCO_3$ for $\xi = 0.76$-0.95[25]), it became clear that such a strong dependence is not observed for γ_1. The experimental data for $CsMnF_3$ and $CsMnCl_3$ are

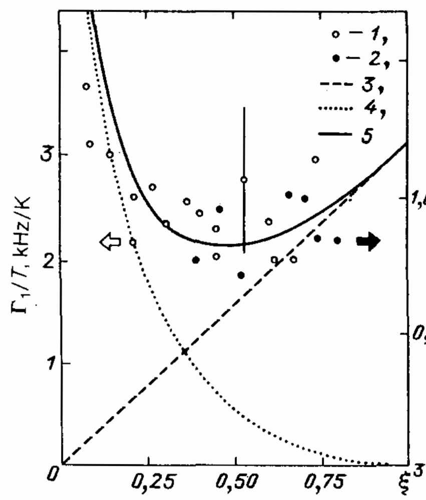

FIG. 13. Frequency dependence of the nuclear spin wave relaxation rate Γ_1 in (ξ^3,Γ/T) coordinates at $\alpha k = 1$ kOe in *1*) $CsMnF_3$ and *2*) $CsMnCl_3$; *3*) is the theoretical dependence of Eq. (5); *4*) is the contribution to nuclear spin wave relaxation determined by their being scattered by sample defects (Eq. (10)) or boundaries (Eq. (11)), or what is introduced from electron spin waves (Eqs. (9) and (12)); and *5*) is the total dependence (the sum of *3*) and *4*)).

presented in Fig. 13 in the Γ_1/T, ξ^3 coordinates. Such a choice of coordinates enables one to detect additions to the relaxation process $\tilde{\gamma}_R$ denoted by the straight dashed line. It is evident that this process enables one to describe the experimental data only at high frequencies ($\xi \to 1$).

Allowance for processes of elastic scattering of nuclear spin waves by defects is one of the possible explanations for such a discrepancy between theory and experiment. An expression for the nuclear spin wave relaxation rate can be written for them (be they a dislocation,[54,55] point defects,[55] or a paramagnetic impurity[56]) in the form

$$\gamma_d = C_d\frac{(1-\xi^2)^2}{\xi}Tk; \qquad (10)$$

here C_d is a coefficient which depends on the parameters of the nuclear spin wave interaction with defects, and is proportional to their concentration. It is evident that, due to the presence of the free parameter C_d, the linear combination $\tilde{\gamma}_R + \gamma_d$ enables one to describe the experimental data for γ_1 over the entire frequency range (the solid curve in Fig. 13 corresponds to the case $C_d = A_R$). However, here each time the magnitude of the coefficient C_d must have a rigorously determined value with $\pm 25\%$ accuracy for the different antiferromagnetics. A random coincidence of the C_d coefficients with values necessary for "smoothing" the frequency dependence of γ_1 for the different samples appears to be of extremely low probability.

The elastic scattering processes for the nuclear magnons at the sample boundaries also give a dependence that is analogous to Eq. (10). For them, the relaxation rate is

$$\gamma_b = \frac{\partial\omega_{nk}}{\partial k}\frac{1}{2L} \propto \frac{(1-\xi^2)^2}{\xi}Tk, \qquad (11)$$

where L is the characteristic size of the crystal. The dotted curve in Fig. 13 can be determined by the γ_b of Eq. (11) if $L \approx 2$ mm, which corresponds to the sizes of the samples investigated. One can interpret in favor of this explanation the results of an experiment[57] on the parametric excitation of electron spin waves in $CsMnF_3$ at low temperatures, where an increase was observed for the relaxation rate of electron magnons with decreasing sample dimensions.

One can suggest another way to explain the frequency dependence of γ_1 based on the hypothesis introduced in Section 5.1 about the effect of electron spin wave relaxation on the damping of nuclear spin waves. It turns out[58] that, in the range of the parameters H and T in which experimental results were presented on the parametric excitation of nuclear spin waves, Woolsey–White processes make the main contribution to electron spin wave relaxation: the elastic scattering of f-magnons by fluctuations of the longitudinal component of the nuclear magnetization[18] (an analog of the Richards process) with the relaxation rate

$$\gamma_{\rm WW} = \frac{I(I+1)}{12\pi}(V_0^{1/3}k)\frac{\omega_{\rm n}^2}{\omega_{ek}}\left(\frac{J_0}{k_{\rm B}\theta_{\rm N}}\right)^2. \qquad (12)$$

If one now transforms this expression according to Eq. (9) into a "transformed" nuclear spin wave relaxation rate and adds it to $\tilde{\gamma}_{\rm R}$, then one obtains the result

$$\gamma_1 = A_{\rm R}[\xi^3 + \frac{1}{\xi}(1-\xi^2)^2]Tk. \qquad (13)$$

This expression describes the experimental data over the entire frequency range (the solid curve in Fig. 13). Here the calculated values $A_{\rm R}^{\rm theo}$ agree well with the $A_{\rm R}^{\rm exp}$ values obtained by the method of least squares. The $A_{\rm R}$ coefficients in units of kHz/(10^5 cm^{-1} K) are shown in Table II.

Thus, allowance for the contributions of the Richards and Woolsey-White processes enables one to describe the experimental data for γ_1 well with dependences on T, k, and ω_{nk} without any kind of fitting parameters.

5.3. The relaxation of nuclear spin waves with a strong temperature dependence $\gamma_2 \propto T^5$

We now go to a discussion of the mechanisms which are responsible for the increase of the nuclear spin wave relaxation rate with an increase of temperature. We define the value of this relaxation γ_2 as the difference between the total measured relaxation rate $\gamma(T,k,\omega_{nk})$ and $\gamma_1(T,k,\omega_{nk})$ extrapolated assuming $\gamma_1 \propto T$ for higher temperatures. The temperature dependence of γ_2 obtained for $CsMnF_3$ is presented in Fig. 14. The experimental results are described well by a law $\gamma_2 \propto T^{5\pm0.5}$. An analogous dependence for the nuclear spin wave relaxation rate was discovered earlier,[23,24,25] in $MnCO_3$ (Fig. 4). As was already noted, the results of a detailed theoretical analysis of possible intrinsic relaxation processes carried out in Ref. 28 showed that only a process of merging a nuclear magnon and a phonon into a phonon (n + ph → ph) enables one to explain a $\gamma_{\rm n} \propto T^5$ dependence in a two-sublattice antiferromagnetic. One must note that, besides the strong temperature dependence, theory[28] also

TABLE II. Theoretical and experimental values of the parameter A_R.

Crystal	$A_{\rm R}^{\rm theo}$	$A_{\rm R}^{\rm exp}$
$CsMnF_3$	2.7 ± 1	3.2 ± 0.8
$CsMnCL_3$	3.0 ± 1.6	1.2 ± 0.3
$MnCO_3$	2.5 ± 1	2.3 ± 0.7

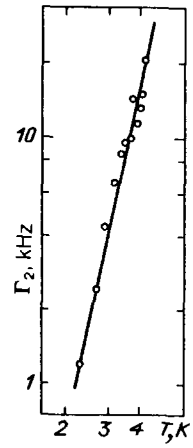

FIG. 14. Temperature dependence of the nuclear spin wave relaxation rate Γ_2 in $CsMnF_3$ at the pumping frequency $\nu_p = 904$ MHz for $\alpha k = 0.9$ kOe, which demonstrates the behavior $\Gamma_2 \propto T^5$.

predicts a strong dependence on frequency $(1-\xi^2)^2$ and the dependence $1/k$ on the wave number. We made suitable measurements of the dependences of γ_2 on these parameters in $CsMnF_3$ and $MnCO_3$. Experimental data for γ_2 at $T = 4.2$ over a broad frequency range are presented in Fig. 15. The coordinates in Fig. 15 have been chosen in such a manner that a straight line passing through the origin of coordinates corresponds to the theoretical dependence $\gamma_{\rm n2ph}(\omega_{\rm nk})$. It is evident that the experimental data for $MnCO_3$ are described well only within a n + ph → ph relaxation process. At the same time, the frequency dependence of γ_2 in $CsMnF_3$ differs significantly from a straight line (Fig. 15). This circumstance stimulated searches for a nuclear spin wave relaxation mechanism with the same temperature dependence as for $\gamma_{\rm n2ph} \propto T^5$, but with a significantly differ-

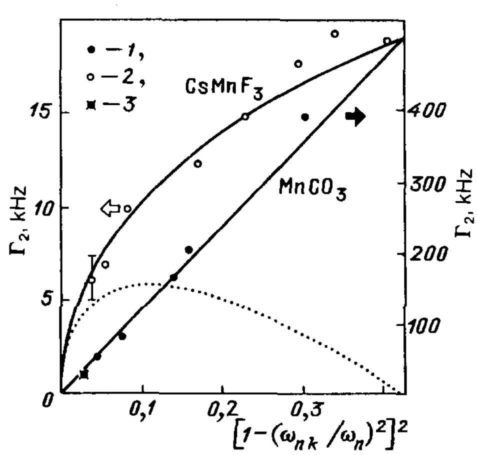

FIG. 15. Frequency dependences of the relaxation rates Γ_2 in 1) two-sublattice $MnCO_3$ antiferromagnetics and 2) six-sublattice $CsMnF_3$ antiferromagnetic for nuclear spin waves with $k = 10^5$ cm^{-1} for $T = 4.2$ K. The straight line passing through the origin of coordinates corresponds to the theoretical frequency dependence of Eq. (4) for a process with two phonons participating, and the dotted curve is for a process with two electron magnons participating (Eq. (14)). The solid curve drawn through the open circles represents the sum of all the dependences, and 3) is a result from Ref. 25, which demonstrates the amount of scatter of the experimental data obtained on different $MnCO_3$ samples.

ent dependence on nuclear spin wave frequency. Such a mechanism must (by itself or in combination with γ_{n2ph}) describe the experimental frequency dependence of $\gamma_2(\omega_{nk})$. A hypothesis has been formulated that the difference in the behavior of $\gamma_2(\omega_{nk})$ is determined by structural features of $MnCO_3$ and $CsMnF_3$, which are two-sublattice and six-sublattice antiferromagnetics, respectively.[59]

Using data from neutron diffraction research for $CsMnF_3$,[33] we made a calculation of the nuclear-spin oscillations within a six-sublattice model for this antiferromagnetic.[50,60] The results of a theoretical analysis showed that the expressions for the spectrum and frequencies of nuclear spin wave relaxation in the six-sublattice model differ from the analogous expressions in the two-sublattice model of the antiferromagnetic, but in view of the relatively small interval between the two unshifted nuclear magnetic resonance frequencies ($\nu_{1n} = 666$ MHz, $\nu_{2n} = 677$ MHz) the difference is $\sim 1\%$. However, it was discovered in a detailed examination that, besides the spin wave interaction processes, which are also inherent to a two-sublattice antiferromagnetic, a new type of anharmonicity arises in a microscopic six-sublattice model; interaction processes with the participation of three magnons only of the quasiferromagnetic (f) branch of the spectrum (accordingly, also the processes of interaction of nuclear spin waves with two quasiferromagnons). Such a difference is determined by the fact that the local symmetry of two nonequivalent positions of Mn^{2+} ions (MnI-D_{3d},MnII-C_{3v}) in an elementary cell is lower than the symmetry of the cell as a whole (D_{6h}). As a result, it turns out that the coordination sums z_I and z_{II} over the nearest neighbors are not the same for different magnetic ions; therefore, as the result of a distortion of the magnetic sublattices due to an external magnetic field, besides the usual three-particle processes of interaction of two f-magnons with a magnon of the quasiantiferromagnetic branch of the spectrum (depending on the difference $z_I - z_{II}$), a new form of three-magnon interactions arises. The calculation of the nuclear spin wave relaxation rate due to the process of merging of a nuclear magnon and a f-magnon into a f-magnon ($n + m \to m$) leads to the following

$$\frac{\gamma_{n2m}}{2\pi} = C_{n2m}(1-\xi^2)^2\,\frac{gH^2}{\alpha k}\left(\frac{T}{\theta_N}\right)^5. \tag{14}$$

It is evident that this equation differs functionally from Eq. (4) by the dependence $\gamma_n \propto H^2$. However, the quantity H is not an independent variable in the (T,k,ω_{nk}) coordinates; therefore, it can be represented as $H = H(T,k,\omega_{nk})$. For example, at fixed T and k, we have $H = H(\omega_{nk})$. The dependence $\gamma_{n2m}(\omega_{nk})$ is shown by the dotted curve in Fig. 15. It is clear that, by itself, the new relaxation process does not enable one to describe the behavior of γ_2 over the entire frequency range, but only in the high frequency range of nuclear spin waves. Thus, the experimental frequency dependence of γ_2 in $CsMnF_3$ can be described only by a combination of the two nuclear spin wave relaxation processes considered above. The solid curve in Fig. 15 corresponds to the sum $\gamma_{n2ph} + \gamma_{n2m}$:

$$\frac{\gamma_{n2ph}+\gamma_{n2m}}{2\pi}[\mathrm{kHz}] = 10^{-2}(1-\xi^2)^2\{3{,}3 + 1{,}7(H[\mathrm{kOe}])^2\}\frac{T[K])^5}{\alpha k\,[\mathrm{kOe}]}. \tag{15}$$

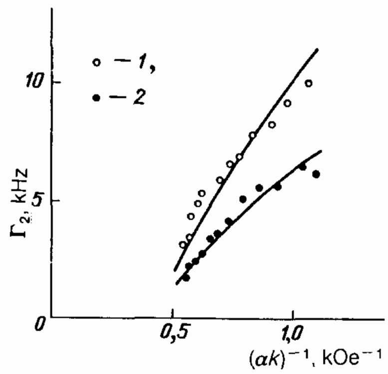

FIG. 16. Dependence of the nuclear spin wave relaxation rate Γ_2 in $CsMnF_3$ on the wave vector magnitude; $\nu_p = 1128$ MHz; *1)* $T = 4.24$ K, and *2)* $T = 3.88$ K. The solid curves have been drawn according to Eq. (15).

The numerical coefficients in Eq. (15) were selected so as to describe the experimental data in the best manner. They correspond to $\tilde{\Theta} = 1{,}100$ K and $C_{n2m} = 0.0037$. One must note that Eq. (15) also describes the experimental dependences $\gamma_2(k)$ well (Fig. 16).

Let us say a few words in connection with the dependence of γ_2 on the magnitude of the wave number in $MnCO_3$. The data of Ref. 25 and the results of our experiments in the $\omega_{nk}/2\pi > 500$ MHz frequency range indicate no dependence of γ_2 on k. An increase of γ_2 with decreasing k is observed at the lowest frequencies; however, this increase is noticeably less than for the $\gamma_{n2ph} \propto 1/k$ dependence. This discrepancy is evidently caused by a not high enough quality of the $MnCO_3$ crystals. Because of lattice defects, quasiparticle interaction processes without the conservation of quasimomentum are possible, and therefore the $1/k$ dependence may be significantly weakened in the $k \lesssim l_d^{-1}$ region (l_d is the characteristic distance between defects). The nuclear spin wave relaxation rate γ_2 in the two-sublattice antiferromagnetic $MnCO_3$ is described well by Eqs. (4) for its dependences on T and ω_{nk} for the $n + ph \to ph$ process with the magnetoelastic constant $\tilde{\Theta} = 1.57 \cdot 10^4$ K.

We now discuss the following question: Do processes of electron spin wave relaxation with a strong temperature dependence (within the hypothesis of relaxation renormalization) exert a noticeable effect on nuclear spin wave damping at $T > 2$ K? From the results of a number of experimental papers that investigate the relaxation of electron magnons in $MnCO_3$[52,61] and $CsMnF_3$,[52,36] it follows that a process with $\gamma_e \propto H^2$ and a strong temperature dependence ($\gamma_e \propto T^6$ to T^8 in the $1.5\ K \leqslant T \leqslant 2.1$ K range) plays a significant role in damping electron spin waves. In its field and temperature dependences, such behavior of the electron spin wave damping is completely described by a three-magnon process,[62] by the merging of two f-magnons into an a-magnon. However, the frequency dependence of electron spin wave damping[63] does not agree with the theory of three-magnon relaxation. Therefore, one may not extrapolate the existing experimental data to the entire range of electron spin wave frequencies connected with the nuclear spin waves being investigated ($\omega_{ek}/2\pi \leqslant 10$ GHz are connected with $\omega_{nk}/2\pi = 400$-600 MHz). We note that an estimate of the contribution under consideration to nuclear spin wave relaxation made from the known damping of an electron spin wave with frequency $\omega_{ek}/2\pi \approx 9$ GHz[12] gives a value much smaller than γ_{n2m}.

5.4. The processes of the interaction of nuclear spin waves with defects

After a detailed investigation of the intrinsic processes of nuclear spin wave relaxation in the antiferromagnetics $MnCO_3$ and $CsMnF_3$, the possibility appeared for studying the damping of these quasiparticles in crystals with defects. The objective of the new investigation consisted of detecting the contribution of extrinsic processes to the observed relaxation rate. The results of such an analysis can be used to estimate the concentration and type of defects in samples being investigated, which usually presents a fairly complicated task.

A non-stoichiometric chemical composition of a crystal, which is unavoidably present to some degree in every sample, is the simplest and most widespread cause of the origin of defects. Therefore a $CsMnF_3$ crystal with intentionally violated stoichiometry was chosen as the object of investigation. The sample contained a 1% excess of manganese compared with the stoichiometric composition; the uniformity of the distribution of the surplus atoms through the volume of the sample was not checked. An x-ray diffraction pattern for the $CsMnF_3$ crystal with violated stoichiometry conformed to the data for pure samples. The results of investigating the nuclear magnetic resonance and antiferromagnetic resonance spectra showed that, within the measurement errors, the frequencies of these resonances and the widths of their lines in the "defective" sample do not differ from the corresponding characteristics of a $CsMnF_3$ sample with nominal stoichiometric composition. The indicated circumstances enable one to state that the difference of the nuclear spin wave relaxation rates in these samples is not connected with significant disruption of the crystal structure or with noticeable changes of the spectra, and is caused only by processes of nuclear spin wave interaction with defects.

The results of the experiments showed that, in the frequency range $\omega_{nk}/2\pi > 400$ MHz, the nuclear spin wave relaxation in a sample with violated stoichiometry remained the same, within the accuracy of the measurement errors, as in a sample of nominal stoichiometric composition: $\gamma = \gamma_1 + \gamma_2$. For a further reduction of the frequency of the nuclear spin waves investigated, a process $\gamma_{1d} \propto Tk$ with a significantly stronger frequency dependence than for the elastic scattering process γ_d of Eq. (10) is observed on the background of the intrinsic processes $\gamma_1 + \gamma_2$.

The frequency dependence of the total nuclear spin wave relaxation $\gamma_1 \propto Tk$ is presented in Fig. 17. It is evident that, as the frequency $\omega_{nk}/2\pi$ is reduced from 400 MHz to 250 MHz, the value of γ_1 increases by approximately 15 times. This circumstance indicates the nature of the "turning on" with decreasing frequency of a new process of relaxation which may be connected either with the impossibility of satisfying conservation laws for this process at the high frequencies $\omega_{nk}/2\pi \gtrsim 400$ MHz, or with a very strong frequency dependence of this process, because of which it starts to appear only at low frequencies.

Besides the process with γ_{1d}, the turning on of one more relaxation process γ_{2d} was observed at lower frequencies

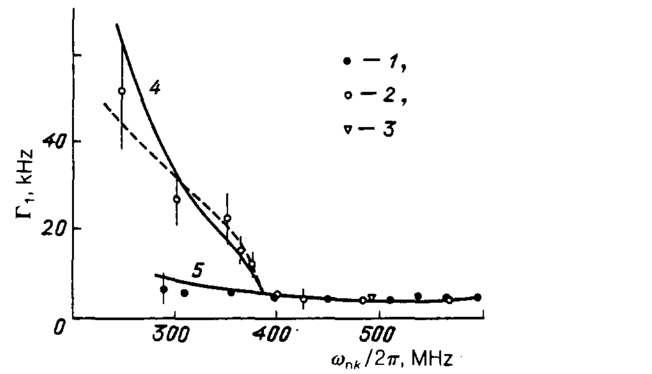

FIG. 17. Frequency dependence of the nuclear spin wave relaxation rate $\Gamma_1 \equiv \gamma_n/2\pi$ in $CsMnF_3$ at $T = 2$ K and $\alpha k = 0.7$ kOe: *1*) A sample of nominal stoichiometric composition; *2*) a sample with violated stoichiometry; *3*) a point from Ref. 16; curve *4* has been drawn according to Eq. (13); curve *5* corresponds to the sum of γ_{nd1} (Eq. (17)) and the intrinsic relaxation of Eq. (13). The theoretical frequency dependence from Ref. 64 is shown by the dashed curve.

$\omega_{nk}/2\pi \leqslant (358 \pm 5)$ MHz. The relaxation rate γ_{2d} is, within the accuracy of the measurement errors, independent of the wave number k, while its temperature dependence presented in Fig. 18 has the form $\gamma_{2d} \propto T^3$. An analogous process of relaxation was observed[51] in a $CsMnCl_3$ crystal (Figs. 19 and 20); therefore, it is natural to suppose that it has the same nature in $CsMnF_3$ and $CsMnCl_3$ and is associated with the relaxation of nuclear spin waves by defects.

We now go to an interpretation of the discovered nuclear spin wave relaxation processes that are determined by their interaction with the defects of the crystal. We emphasize that not one of the mechanisms for nuclear spin wave relaxation by crystal defects suggested in Refs. 54, 55, and 56 enables one to describe the $\gamma_{1d}(\omega_{nk})$ or $\gamma_{2d}(\omega_{nk})$ dependences. Therefore, one must seek an explanation of the experimental data by invoking new processes for the interaction of nuclear magnons with defects that have an internal structure.

We consider the simplest model for such a defect, a two-level system with the transition frequency ω_d. It is natural to suppose that the anomalies in the nuclear spin wave relaxation are associated with ω_d, and specifically either with

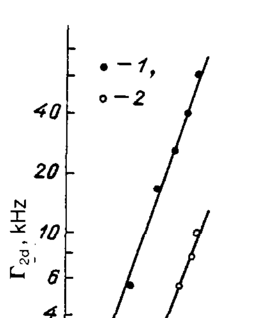

FIG. 18. Temperature dependences of the relaxation rate Γ_{2d} *1*) in $CsMnF_3$ at $\nu_p = 706$ MHz and *2*) in $CsMnCl_3$ at $\nu_p = 896$ MHz.

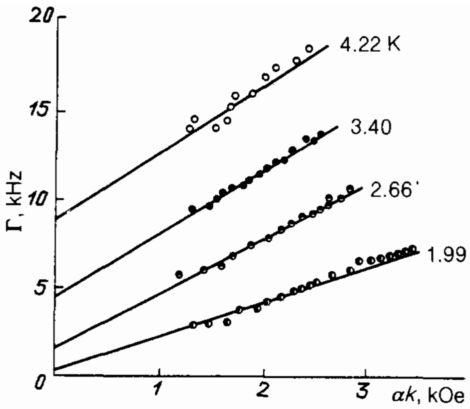

FIG. 19. The nuclear spin wave relaxation rate in $CsMnCl_3$ at $\frac{1}{2}\nu_p = 475$ MHz.[51]

$\omega_d/2\pi \simeq 400$ MHz ($CsMnF_3$) and 480 MHz ($CsMnCl_3$), or with double these frequencies. Since, for the first assumption, features must arise in the nuclear spin wave spectrum (the pushing apart of the nuclear spin wave branches and of the local level) which are not observed experimentally, therefore from now on we shall discuss only the second possibility. Two types of relaxation processes are most significant in this case: a) the process of merging of two nuclear magnons with the excitation of a transition in the two-level system $\omega_{nk} + \omega_{nq} = \omega_d$, and b) a process of "slow" nuclear spin wave relaxation in this two-level system. A process of the first type which, in principle, enables one to describe γ_{1d} in terms of dependences on T, k, and ω_{nk} (the dashed curve in Fig. 17) was considered in Ref. 64. However, to describe the quantity γ_{1d}, one must assume that the characteristic energy binding a defect with the magnetic matrix is extremely large ($\lambda \sim 15$ K). Let us consider the other variant for interpreting the γ_{1d} and γ_{2d} processes by means of the theory of slow nuclear spin wave relaxation. A quantum theory for the slow relaxation of spin waves (damping determined by modulation of the distance between the levels of a rapidly relaxing two-level system) has been constructed by Mikhaĭlov and Farzetdinova.[65] In the case of interest to us, one can

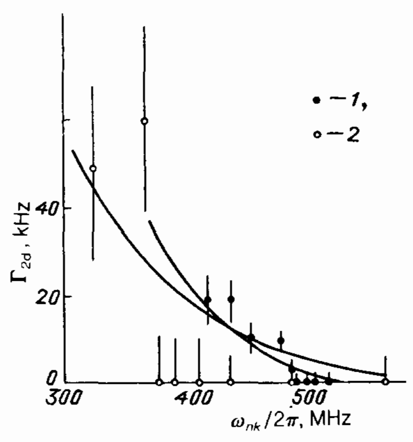

FIG. 20. Frequency dependence of the nuclear spin wave relaxation rate at $T = 4.2$ K which corresponds to a process with $\Gamma_{2d} \propto T^3$. *1*) for $CsMnCl_3$; *2*) for $CsMnF_3$ with non-stoichiometric composition; the curves have been drawn according to Eq. (18).

write the equation for slow nuclear spin wave relaxation in the form

$$\gamma_{nd} = \frac{n_d}{8}\frac{|\Phi|^2}{\hbar\omega_{nk}k_BT}[\gamma_\parallel(\omega_d + \omega_{nk}) + \gamma_\parallel(\omega_d - \omega_{nk})]; \qquad (16)$$

here n_d is the concentration of defects, $\Phi = \lambda_1(1-\xi^2)(k_BT/\xi\hbar\omega_n)^{1/2}$ is the amplitude of the process, $\lambda_{1,2}$ are the characteristic energies binding a defect with the magnetic matrix, and $\gamma_\parallel(\Omega)$ is the relaxation rate for a two-level system at the frequency Ω. The longitudinal relaxation $\gamma_\parallel(\Omega)$ can be determined by:

1) direct processes (a transition from an upper to a lower level with the generation of a nuclear magnon with frequency $\omega_{nq} = \Omega$), and

2) processes of merging of an "excited defect" and an electron spin wave with frequency ω_{ek} into an electron magnon $\omega_{ek} + \Omega$. It is easy to understand that the first process is permitted if the frequency Ω falls into the nuclear spin wave zone, which is possible only for $\omega_d - \omega_{nk} = \omega_{nq}$ (i.e., $\omega_{nk} \leqslant \omega_d - \omega_{n0}$). It is interesting to note that the last condition, which is written from the law of conservation of energy for this relaxation process, agrees exactly with the analogous condition in Ref. 64 for the process of merging two nuclear magnons with the excitation of a defect.

Calculation of the direct processes leads to the following result:[66]

$$\gamma_{nd_1} = \frac{3n_d}{2^5\pi I(I+1)}\frac{(\lambda_1\lambda_2)^2 J_0 T}{(\hbar\omega_n)^3 k_B^2\theta_N^3}\frac{(1-\xi^2)^2}{\xi^2(\xi_d - \xi)}g\alpha\bar{k}, \qquad (17)$$

where

$$(\alpha\bar{k})^2 = H_\Delta^2\left[\frac{(\xi_d - \xi)^2}{1-(\xi_d-\xi)^2} - \frac{\xi^2}{1-\xi^2}\right] + (\alpha k)^2.$$

It is evident that $\gamma_{nd_1} \propto T$, and the $\gamma_{nd_1}(k)$ dependence in the frequency range $\omega_{nk} \sim \frac{1}{2}\omega_d$ is close to linear, which agrees with the experimental data for $\Gamma_{1d}(T,k)$. The thick curve in Fig. 17 corresponds to the sum of Expressions (13) and (17). It is evident from Fig. 17 that the frequency dependence of γ_{nd_1} also satisfactorily describes the experimental data. Here the fitting parameters equal $\omega_d = 2\pi\cdot 732$ MHz and $n_d^{1/2}\lambda_1\lambda_2 = 0.013$ K^2, i.e., for $n_d \sim 0.01$ and $\lambda_1 \sim \lambda_2$, we have $\lambda_1 \sim \lambda_2 \sim 0.4$ K. These are entirely reasonable values for the energy binding a defect with a magnetic matrix.

We now consider the second process—the merging of an electron spin wave with an excitation of a two-level system into a new electron spin wave $\omega_{eq_2} = \Omega + \omega_{eq_1}$. We note that for processes of this type, the prohibitions of energy conservation laws (as in the first process) are not imposed. The calculation gives[66]

$$\gamma_{nd_2} = \frac{n_d}{2^3\pi I(I+1)}\frac{(\lambda\lambda_1)^2 J_0^2 T^3}{\hbar^3\omega_n^2 k_B^3\theta_N^6}\frac{(1-\xi^2)^2}{\xi^2}. \qquad (18)$$

Thus, $\gamma_{nd_2} \propto T^3$ is independent of k, which agrees with the experimental results for Γ_{2d}. The $\gamma_{nd_2}(\omega_{nk})$ frequency dependences are shown by the solid curves in Fig. 20. The theoretical curve for $CsMnF_3$ has been drawn using the results of reducing the relaxation Γ_{1d} according to Eq. (17). The value of λ_1 obtained from this, and also the known value $n_d = 0.01$ were substituted into Expression (18) (it was assumed here that $\lambda \sim \lambda_1$). It is evident that the theoretical curve of $\gamma_{nd_2}(\omega_{nk})$ as a whole correctly describes the quanti-

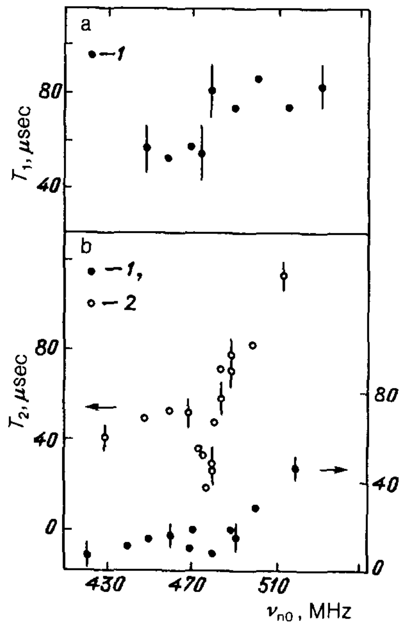

FIG. 21. Frequency dependence of the times for nuclear a) spin-lattice and b) spin-spin relaxation in $CsMnCl_3$ at the temperatures *1*) 4.2 K and *2*) 1.6 K.[51]

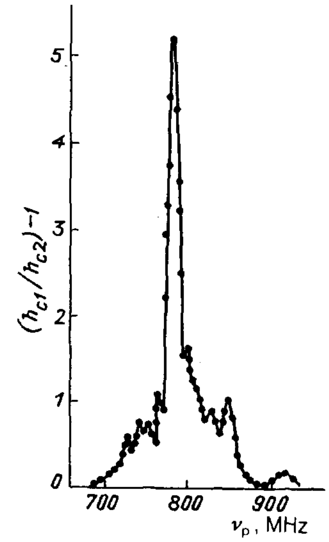

FIG. 22. Dependence of the hardness parameter in $CsMnF_3$ on pumping frequency at $T = 1.86$ K and $H = 0.8$ kOe.

ty Γ_{2d} and the variation of the frequency dependence, although a noticeable discrepancy between theory and the experimental data is also observed in the region of the hardness peak (see Section 6).

For $CsMnCl_3$ Expression (18) describes well the dependence of Γ_{2d} on all parameters. The frequency dependence of γ_{nd_2} with the value of the parameter $n_d^{1/2}\lambda\lambda_1 = 0.021$ K^2 is shown in Fig. 20. In $CsMnCl_3$, along with experiments on the parallel pumping of nuclear spin waves, investigations were also conducted on the relaxation characteristics of the nuclear subsystem by the nuclear spin echo method. The results of measuring the times of the nuclear spin-lattice relaxation T_1 and of the damping of the coupled nucleus-electron precession are presented in Fig. 21. It is evident that near the frequency $\omega_{n0} \simeq 2\pi\cdot 480$ MHz, which corresponds to an increase of γ_{2d}, features in the behavior of T_1 and T_2 are observed, and furthermore, the first one has the form of a step, and the second one that of a resonance minimum whose position and relative depth are independent of temperature, and its width decreases noticeably with decreasing temperature. These results evidently indicate that the frequency $\omega_d/2\pi$ in $CsMnCl_3$ equals or is a multiple of the frequency 480 MHz.

6. FREQUENCY RESONANCE FEATURES OF THE PARAMETRIC EXCITATION OF NUCLEAR SPIN WAVES IN $CsMnF_3$

As is well known, the parametric excitation of electron spin waves in all antiferromagnetics of interest to us at liquid helium temperatures has a rigid character. The start and ending of the parametric process occurs at different values of a magnetic microwave field on a sample h_{c1} and h_{c2} ($h_{c1} > h_{c2}$), respectively.[36,61,67] We detected this effect during the investigation of nuclear spin waves only in 1982 for $CsMnF_3$ samples,[50] i.e., 13 years after the first observation of paramagnetic nuclear spin waves. Actually, the rigid character of nuclear spin wave excitation is observed in a comparatively narrow range of frequencies $\nu_p = 700$-900 MHz, and furthermore, the frequency dependence of the hardness parameter $\zeta = (h_{c1}/h_{c2}) - 1$ has a clearly pronounced resonance character (Fig. 22). The value of ζ for the hardness peak changes by severalfold from sample to sample, but the position of the peak remains constant for all the samples, and is independent of temperature in the range from 1.5 K to 4.2 K, and of the magnetic field with ± 2 MHz accuracy. The value of ζ for the peak increases with decreasing temperature, for example, in a sample with maximum hardness, from 3.6 at $T = 4.2$ K to 7 at $T = 1.8$ K. However, if one considers here the switching-off part of the relaxation as the hardness parameter, as was done in Ref. 61, i.e., $\Delta\Gamma = \Gamma(h_{c1}) - \Gamma(h_{c2})$, then this value for the hardness peak increases almost linearly with T.

A number of interesting features of the behavior of nuclear spin waves is observed in the region of the hardness peak. First of all, there is the peak in nuclear spin wave relaxation which was mentioned in Section 5.1, and which supposedly is determined by a feature in the relaxation of electron spin waves at the point of their intersection with the longitudinal sound branch (Fig. 23). As is evident from Fig. 23, the peak for nuclear spin wave relaxation is a maximum at the frequency $\nu_p \approx 775$ MHz and decreases rapidly as the pumping frequency changes. The electron magnons coupled to these nuclear spin waves have the frequency $\nu_{ek} \approx 7$ GHz, for which experimental research has not been conducted; therefore, it does not appear to be possible to verify Eq. (9) for the relaxation introduced. However, for the condition that Expression (9) is valid, the peak in the electron spin wave relaxation must be approximately an order of magnitude larger than the analogous peak for electron spin waves with high ($\nu_{ek} \approx 18$ GHz) frequencies.

Besides the "magnon-phonon" peak ($H = 0.8$-1.0 kOe in Fig. 23) near the limiting field for nuclear spin wave pumping, a step is observed which approximately corresponds to $\tilde{q} \approx \frac{1}{2}\bar{k} \simeq 5\cdot 10^4$ cm^{-1}. Since the quantity $\tilde{q}$ is practi-

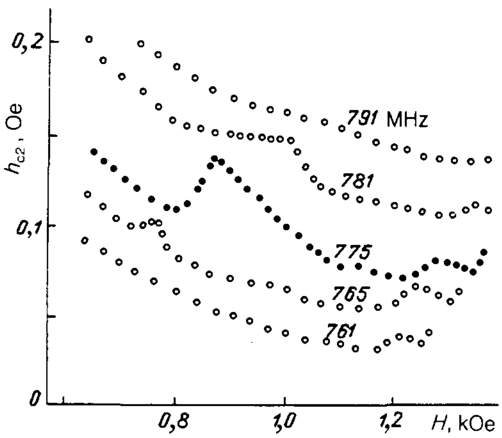

FIG. 23. Dependence of the threshold field h_{c2} on H in $CsMnF_3$ at $T = 1.86$ K and different pumping frequencies. The scale along the ordinate corresponds to the curve for $\nu_p = 775$ MHz. The remaining curves have been parallel shifted for clearness without changing the scale, since the h_{c2} values at all frequencies coincide with $\pm$ 10% accuracy.

cally independent of the nuclear spin wave frequency and temperature, one can suppose that this step is determined by a dimensional effect with the characteristic length $l = 2\pi/\tilde{q} = 1.2\cdot 10^{-4}$ cm. It is possible that the intensification of the magnon-phonon peak is connected with this effect, since it is observed for the nuclear spin wavelength $\lambda = \frac{1}{2} l$, and furthermore, the magnon-phonon peak has an asymmetric shape, and its shape is distorted differently for the $\lambda > \frac{1}{2} l$ or $\lambda < \frac{1}{2} l$ cases.

The parametric excitation of electron spin waves in $CsMnF_3$ has one more outstanding feature—a sharp hexagonal anisotropy of the threshold h_c for the parametric excitation upon rotation of the external field H in the basal plane of the crystal.[68] Apparently this anisotropy is determined by the dependence of the electron spin wave relaxation rate on the direction of the magnetic field in the basal plane. In this case, according to the damping renormalization hypothesis, the contribution of the electron spin waves to nuclear spin wave relaxation must also depend on the direction of H. As was also expected, anisotropy of both thresholds was also observed during the excitation of nuclear spin waves (Fig. 24), and furthermore, the amount of this anisotropy corresponds to the angular dependence of the nuclear spin wave relaxation calculated according to Eq. (9). One more interesting effect was discovered during the investigation of the angular dependences of the threshold fields for nuclear spin wave excitation—the intensification of the magnon-phonon peak is strongly anisotropic, and it almost disappears at the $h_c(\varphi)$ maximum. One common property is characteristic for all the indicated phenomena: the hardness value, the amplitude of the magnon-phonon peak, the degree of threshold anisotropy, and the effect of intensification of the longitudinal magnon-phonon peak change from sample to sample. This indicates that the enumerated phenomena are not inherent to an ideal crystal, but are determined by the defects of a sample.

In our opinion, all the enumerated anomalies may be of the nature of dislocations. One may cite the following arguments in favor of such a conclusion. The hexagonal anisotropy of a threshold evidently means that the magnon relaxation rate depends on the direction of the wave vectors k.[68] At the same time, the dislocations have a tendency to line up along definite crystallographic directions and can, in principle, lead to anisotropy of h_c. The anisotropy of the intensification of the magnon-phonon peak may also be connected with the alignment of the dislocations. The range of frequencies in which the anomalies in nuclear spin wave relaxation being discussed are observed and the size $l \sim 10^{-4}$ cm are also characteristic of the dislocations. Finally, in investigating the nuclear spin echo in $CsMnF_3$, it was shown in Ref. 41 (Fig. 25) that the dislocation contribution to nuclear spin wave relaxation with $k \lesssim 10^4$ cm^{-1} may have a noticeable value. The interaction of magnons with dislocations might explain all the anomalies considered in nuclear spin wave relaxation. However, the theory for such interaction is lacking at present, and the experimental data are apparently insufficient for drawing any kind of final conclusions about the nature of the phenomena enumerated in this section.

CONCLUSION

As a result of the comprehensive experimental and theoretical research conducted in recent years, the main mechanisms of nuclear spin wave interaction in antiferromagnetics have been determined, and the validity of using

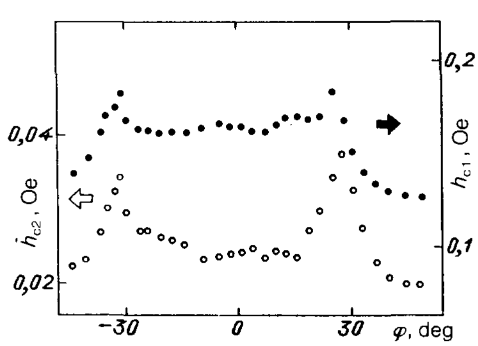

FIG. 24. Dependences of the threshold fields h_{c1} and h_{c2} for the parametric excitation of nuclear spin waves in $CsMnF_3$ on the direction of H in the basal plane of a crystal ($\varphi = 0$ corresponds to the binary axis of the crystal). $T = 1.95$ K, $H = 0.94$ kOe, and $\nu_p = 784$ MHz.

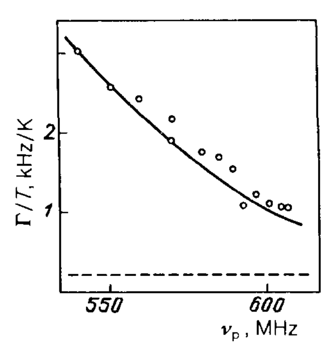

FIG. 25. Frequency dependence of the relaxation rate for the coupled nucleus-electron precession that is excited in $CsMnF_3$ by the parametric echo method;[41] the theoretical $\tilde{\gamma}_R$ dependence calculated according to Eq. (13) is indicated by the dashed line; the solid curve corresponds to the sum of γ_R and of the contribution of the dislocations[54,55] to the damping of the spin echo signal.

the threshold equation for the nn- and en-processes has been demonstrated.[69]

Progress in studying the spectrum and relaxation of nuclear spin waves was achieved because of the high accuracy in recording the threshold h_c of paramagnetic resonance by a modulation method. This enabled one to conduct measurements of h_c in three antiferromagnetics, $MnCO_3$, $CsMnF_3$, and $CsMnCl_3$, over fairly broad ranges of temperatures, magnetic fields, and pumping frequencies. To explain the series of experimental dependences $\gamma_n(T,H,\omega_p)$ obtained, a theoretical model of nuclear spin wave relaxation adequate for them was suggested, which basically describes the experimental regularities for all the parameters.

It turned out that one can describe the damping of nuclear magnons in the three antiferromagnetics indicated above at relatively low temperatures ($T \lesssim 2$ K) without any kind of fitting parameters by processes of scattering of spin waves by fluctuations of the magnetization of the nuclear subsystem. Here in the high frequency range, the Richards process, the scattering of nuclear spin waves by the fluctuations, makes the main contribution to relaxation. According to the hypothesis we have suggested, the contribution of the process introduced from the electron spin wave branch of electron magnon scattering by the same fluctuations becomes the main one at low frequencies. The presence of this introduced damping and the value of the renormalization coefficient have been verified experimentally from the electron spin wave relaxation anomalies transformed to nuclear spin wave relaxation.[7)]

Other processes also become significant in the behavior of nuclear spin wave relaxation at higher temperatures ($T > 2$ K). Thus, in $MnCO_3$, the nuclear spin wave relaxation rate is determined by a magnon-phonon process—by the merging of a nuclear magnon and a phonon into a phonon. In $CsMnF_3$, the process of merging a nuclear magnon and an electron magnon into an electron magnon exists along with the magnon-phonon process. We note that the magnon-magnon process is permitted only in multi-sublattice antiferromagnetics.

Besides the intrinsic channels for nuclear spin wave relaxation, we have experimentally detected for the first time extrinsic processes determined by crystal defects, and have suggested a theoretical model for the interaction of nuclear spin waves with the defects that enables us to describe the main results satisfactorily.

It is important to note that, by comparison with the electron spin waves, the system of nuclear spin waves differs in its low heat capacity and is weakly coupled to the lattice. This circumstance enables one to attain extremely high levels of parametric excitation for nuclear magnons and to observe phenomena determined by strong nonlinearities. Thus, a nuclear spin wave system was investigated in the steady-state and self-oscillation (periodic and chaotic) regimes of absorbing microwave power in Refs. 72–78. A double parametric resonance of nuclear magnons[46,76] and a process for generating elastic oscillations of a sample[75,77] have been studied in detail. An effect of weak signal amplification near the bifurcation of cycle generation has been detected for the first time.[77] A discussion of the physical results obtained beyond the threshold of parametric excitation of nuclear spin waves is beyond the scope of this review.

Thus, the problem of the kinetics of slightly nonequilibrium excitations in a system of coupled nucleus-electron oscillations of an antiferromagnetic in the range of liquid helium temperatures has been solved in its general features.[8)] The information obtained serves as a good basis for the detailed study of the strongly nonequilibrium phenomena in this convenient model system.

[1)] The name reflects the geometry for exciting homogeneous oscillations by a magnetic microwave field: $\mathbf{h}\perp\mathbf{H}$ (the f-mode), and $\mathbf{h}\|\mathbf{H}$ (the a-mode).

[2)] Subsequently the threshold equation was made more precise in Refs. 14 and 15.

[3)] This idea was used in a theoretical paper by Soares and Rezende.[19]

[4)] Note that the idea of phase space separation into regions with strong and weak departures of the nuclear spin wave frequency ω_{nk} from ω_n was considered earlier by Turov and Petrov.[29] However, they supposed that the cutoff wave number k_0, up to which the concept of waves in a nuclear subsystem has meaning, is determined by the radius r_{SN} of an indirect Suhl-Nakamura interaction ($k_0 \sim r_{SN}^{-1}$). In comparing k_0 and k_*, we find $k_0 \ll k_*$.

[5)] In these experiments the frequency ($\omega_m/2\pi = 100$ kHz) and amplitude ($H_m < 0.07$ Oe) were chosen so that the effect of modulation on the pumping threshold was negligibly small.

[6)] It is interesting to note that a relation similar to Eqs. (9) (without derivation) was used in Ref. 22 for modes with $k = 0$ to calculate the width of a nuclear magnetic resonance line. A satisfactory description of the experimental results was obtained here.

[7)] It is interesting to note that the hypothesis of damping renormalization in the coupled magnetoelastic waves of the antiferromagnetic $FeBO_3$ enables one to explain the field dependence of the phonon relaxation rate ($\propto H^{1/2}$; see Ref. 70) as a result of a three-magnon electron spin wave relaxation process introduced from the electron magnetic subsystem.[71]

[8)] The development of nuclear spin wave theory for the very low temperature region has been carried out in Refs. 32 and 79. We also note that the whole scope of questions on the physics of highly excited coupled nucleus electron precession (i.e., nuclear spin waves with $k = 0$) is discussed in the monograph of Ref. 80 which appeared recently.

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Translated by Frederick R. West