

# Experimental investigation of a system of Mn<sup>55</sup> nuclear spins in antiferromagnetic CaMnCl<sub>3</sub>

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(Submitted 27 August 1982)

Zh. Eksp. Teor. Fiz. **84**, 823–832 (February 1983)

Spin echo and parallel microwave pumping in a wide range of frequencies (400–850 MHz) and temperatures (1.6–4.2 K) are used to investigate the NMR spectrum, the spin-lattice relaxation time ( $T_1$ ) and the spin-spin relaxation time ( $T_2$ ) of Mn<sup>2+</sup>, as well as the relaxation rate  $\Gamma$  of the nuclear spin waves. It is shown that the spectrum consists of two branches corresponding to two nonequivalent crystallographic positions of the Mn<sup>2+</sup> ion, with unshifted frequencies  $\nu_\alpha = 584$  MHz and  $\nu_\beta = 554$  MHz. The dynamic shift of the NMR frequency is calculated for this case, the coupling constant for the electron and nuclear subsystems is found to be  $\Delta^2 = 10.5/T$  kOe<sup>2</sup>/K, and the inhomogeneous exchange constant  $\alpha = 1.25 \times 10^{-5}$  kOe·cm was obtained. An anomalous behavior of all the relaxation processes is observed near 480 MHz and its possible causes are discussed.

PACS numbers: 76.60.Es, 76.60.Lz, 75.50.Ee

## INTRODUCTION

The CsMnCl<sub>3</sub> crystals were synthesized relatively recently.<sup>1</sup> Their structure can be described by a hexagonal crystal lattice with parameters  $a_H = 7.288$  Å,  $c_H = 27.44$  Å or by a rhombohedral lattice with parameters  $a_R = 10.7$  Å,  $\alpha = 42^\circ 26'$  (Refs. 2 and 3) (space group  $D_{3d}^5$ ). It is known from neutron-diffraction investigations<sup>2</sup> that at a temperature  $T < T_N = 67$  K CsMnCl<sub>3</sub> is an easy-plane antiferromagnet in which the ferromagnetic planes are perpendicular to the high-order axis, and the magnetic unit cell is obtained by doubling the crystallographic one and contains eighteen Mn<sup>2+</sup> ions.

The AFMR spectrum and the relaxation of the electron magnons in CsMnCl<sub>3</sub> were investigated in detail in Ref. 4. It was shown that the lower branch of the magnon spectrum is described by the usual expression

$$(\Omega_k/\gamma_e)^2 = H^2 + \alpha^2 k^2 + \Delta^2 \equiv (\Omega_k'/\gamma_e)^2 + \Delta^2, \quad (1)$$

where  $\Omega_k$  and  $k$  are the frequency and wave vector of the magnon,  $H$  is the external magnetic field,  $\gamma_e = 2.8$  GHz/kOe is the electron gyromagnetic ratio,  $\Delta = (11.4/T)^{1/2}$  kOe/K<sup>1/2</sup> is the spectrum gap due to the hyperfine interaction, and  $\alpha$  is the inhomogeneous exchange constant. In addition, the measured<sup>4</sup> static susceptibility was found to be  $\chi_1 = 2.0 \times 10^{-2}$  cgs emu/mol, and this value was used to calculate the exchange field  $H_E = 700 \pm 70$  kOe.

Measurements<sup>4</sup> of magnon damping in CsMnCl<sub>3</sub> have shown that, unlike in other investigated easy-plane antiferromagnets (CsMnF<sub>3</sub>, MnCO<sub>3</sub>),<sup>5,6</sup> here the contribution of the intrinsic relaxation processes is small even at  $T \lesssim 3$  K compared with magnon scattering by impurities and defects of the sample.

The results of AFMR investigations lead us thus to expect in this substance a large dynamic shift of the NMR frequency even at  $T \approx 4$  K. The dynamic shift of the NMR is due to indirect (Suhl-Nakamura) interaction of spins with one another via the magnetically ordered electron spin sys-

tem. In this case there arise collective oscillations of the nuclear spins with spatial dispersion, namely nuclear spin waves (NSW) whose spectrum takes in the simplest case the form

$$\nu_k^2 = \nu_n^2 \Omega_k'^2 / (\Omega_k'^2 + \gamma_e^2 \Delta^2), \quad (2)$$

where  $\nu_n = \gamma_n AM$  is the unshifted NMR frequency;  $\Delta^2 = 2H_E A \langle m \rangle$ ;  $A$  is the hyperfine interaction constant,  $\gamma_n$  and  $\gamma_e$  are the nuclear and electron gyromagnetic ratios;  $M$  and  $\langle m \rangle$  are the mean electronic and nuclear magnetizations of each sublattice.

Since the system of nuclear spins of Mn in CsMnCl<sub>3</sub> was not investigated at all before, we have carried out an exhaustive study with the hope of obtaining the maximum possible information on its spectrum and relaxation parameters. NMR procedures make it possible to study the NSW spectrum at  $k \approx 0$  and determine with high accuracy the pertinent constants; in addition, they yield information on the interactions of the nuclear system with the lattice (the spin-lattice relaxation  $T_1$ ) as well as on the interactions inside the nuclear subsystem itself (the spin-lattice relaxation time  $T_2$ ). However, the relaxation parameter measured by this method is averaged over the entire width of the excited packet ( $\Delta k \approx k \lesssim 10^4$  cm<sup>-1</sup>).

Using the method parallel microwave pumping it is possible to measure NSW relaxation rate  $\Gamma = (4\pi T_2)^{-1}$  with a definite value of the wave number  $k = 10^4$ – $10^6$  cm<sup>-1</sup>, inasmuch as narrow wave packets ( $\Delta k \ll k$ ) are excited in the case of parametric resonance.

All the measurements were made on single-crystal samples grown and kindly furnished by B. V. Beznosikov (Physics Institute, Siberian Division of the USSR Academy of Sciences, Krasnoyarsk). The orientation of the samples was determined by x-ray diffraction; the external and microwave magnetic fields were located in all experiments in the basal plane of the crystal ( $\mathbf{H}, \mathbf{h} \perp \mathbf{C}_3$ ).

## NMR SPECTRA

Since the  $Mn^{2+}$  ions occupy in  $CsMnCl_3$  two nonequivalent positions that differ, generally speaking, in the values of the unshifted NMR frequencies ( $\nu_\alpha > \nu_\beta$ ), the spectrum of the high-frequency branch of the NMR should break up<sup>7</sup> into two regions—the in-phase oscillations of the Mn spins in both positions ( $\nu_1 < \nu_\beta$ ) and antiphase oscillations ( $\nu_\alpha > \nu_2 > \nu_\beta$ ). Therefore the NMR spectrum is determined by four constants  $\Delta_\alpha$ ,  $\Delta_\beta$ ,  $\nu_\alpha$ , and  $\nu_\beta$ , an exact determination of which calls for measurement in both regions. The position of the NMR of Mn in the region of the in-phase oscillations was determined by spin echo in the frequency interval 533–389 MHz and at temperatures 4.2–1.6 K. To observe the spin-echo signal we used a pulsed NMR spectrometer similar to that described in Ref. 8. In these measurements the constant and microwave magnetic fields were mutually perpendicular. The spin echo in  $CsMnCl_3$  behaves mainly as in other substances with dynamic shift of the NMR frequency, but reveals a large number (about 10) of intense additional echo signals at  $t = 3\tau, 4\tau, \dots$  ( $\tau$  is the distance between the exciting pulses). The NMR spectrum in the region of the in-phase oscillations of the Mn spins at  $T = 4.2$  K is shown in Fig. 1.

In the region of the antiphase oscillations, the NMR was observed by saturating the line with an additional microwave oscillator. The observation procedure is the following. The measurement circuit is first tuned in such a way that spin echo is observed (in the in-phase oscillation region) at a definite field value  $H$ . A microwave field from the additional cw oscillator, with frequency  $f > \nu_\beta$ , is then applied to the sample. If this frequency coincides with the frequency of the antiphase oscillations of the Mn spins, absorption of micro-

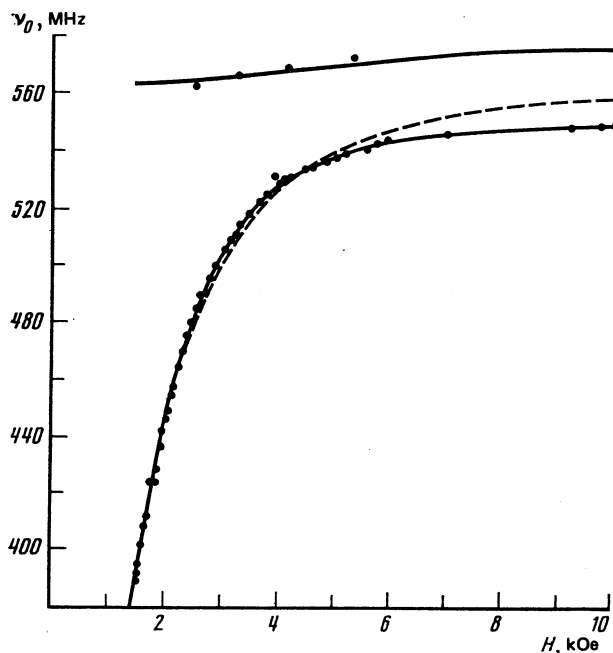


FIG. 1. NMR spectrum for  $Mn^{55}$  in  $CsMnCl_3$  at  $T = 4.2$  K. Solid curve—calculation by Eq. (3), dashed—by (4).

wave power by the samples causes the nuclear-spin system to overheat, the frequency of the in-phase oscillations is shifted, and the echo signal vanishes. At low powers of the additional oscillator absorption (and accordingly, overheating) is observed only within the NMR line width of the antiphase oscillations ( $\Delta\nu \approx 0.5$  MHz), and when the power is increased the NMR line is captured and overheating is observed in a certain frequency range between  $\nu_2$  and  $\nu_\alpha$ . The maximum frequency at which the capture is observed yields the lower-bound estimate  $\nu_\alpha = 583$  MHz.

Calculation of the dynamic shift of the NMR frequency in the presence of two unshifted oscillation frequencies was carried out by Welsh<sup>7</sup> only for the case of a small dynamic shift. The smallness criterion ( $\Delta^2/T \ll H^2$ ) is not satisfied in practice in the greater part of the range of the magnetic fields employed. We have therefore recalculated the dynamic shift of the NMR frequency without using the small-shift approximation. The resultant NMR spectrum consists of two branches that are the roots of a biquadratic equation with the following coefficients:

$$\nu_1^{-2} + \nu_2^{-2} = \nu_\alpha^{-2} (1 + \gamma_e^2 \rho_\alpha \Delta_\alpha^2 / \Omega_0'^2) + \nu_\beta^{-2} (1 + \gamma_e^2 \rho_\beta \Delta_\beta^2 / \Omega_0'^2),$$

$$(\nu_1 \nu_2)^{-2} = (\nu_\alpha \nu_\beta)^{-2} [1 + \gamma_e^2 (\rho_\alpha \Delta_\alpha^2 + \rho_\beta \Delta_\beta^2) / \Omega_0'^2], \quad (3)$$

this being a natural generalization of Welsh's results.<sup>7</sup> Here  $\Omega_0'$  is the corresponding AFMR branch;  $\rho_{\alpha(\beta)}$  is the fraction of the atoms in the position  $\alpha(\beta)$  and  $\Delta_{\alpha(\beta)}^2 = 2H_E A_{\alpha(\beta)} \langle m_{\alpha(\beta)} \rangle$ .

Although it is not difficult to calculate from (3) the analytic form of the NMR spectrum, the resultant equations are quite unwieldy. We consider therefore the asymptotic behavior in strong and weak magnetic fields. The NMR spectrum consists of two branches, of which the lower (in-phase) approaches asymptotically  $\nu_\beta$  in strong fields, while in weak fields (relative to  $\Delta$ ) it is described by the equation

$$\nu_1^2 = \nu_{\alpha\beta}^2 \Omega_0'^2 / (\Omega_0'^2 + \gamma_e^2 \Delta_{\alpha\beta}^2),$$

$$\nu_{\alpha\beta}^2 = 2\nu_\alpha^2 \nu_\beta^2 / (\nu_\alpha^2 + \nu_\beta^2), \quad \Delta_{\alpha\beta} = \rho_\alpha (\nu_\alpha \nu_\beta^2 / \nu_\alpha^2) \Delta_\alpha + \rho_\beta (\nu_\alpha \nu_\beta^2 / \nu_\beta^2) \Delta_\beta, \quad (4)$$

which is similar in form to the usual formula for the dynamic shift of the NMR frequency. The main difference is that the coefficient  $\nu_{\alpha\beta}$  with dimension of frequency does not coincide with the unshifted NMR frequency  $\nu_\beta$ . The temperature dependence of the NMR spectrum is determined by the relation  $\Delta^2 \propto \langle m \rangle \propto T^{-1}$  and corresponds to the usual dynamic frequency shift (this was verified in experiment).

The branch  $\nu_2$  of the antiphase oscillations of the nuclear spins depends weakly on the magnetic field and on the temperature. In strong magnetic field it approaches  $\nu_\alpha$  asymptotically, and in weak fields it tends to the value

$$\nu_2(0) = \left( \frac{\nu_\alpha^2 + \nu_\beta^2}{2} \right)^{1/2} + \frac{\rho_\beta - \rho_\alpha}{|\rho_\beta - \rho_\alpha|} O(|\nu_\alpha - \nu_\beta|). \quad (5)$$

The quantities most accurately determined in experiment are  $\nu_\alpha$  and  $\nu_\beta$  (from measurements in strong fields and at large amplitudes of the microwave field). It follows from our results that in  $CsMnCl_3$  we have  $\nu_\alpha = 584 \pm 1$  MHz and

$\nu_\beta = 554 \pm 1$  MHz. From the value  $\nu_2(0) \approx 564$  MHz we can determine next the sign of the second term of (5), and consequently establish the correspondence between the hyperfine frequencies and the positions of the  $\text{Mn}^{2+}$  ions in the crystal lattice. It was found that  $\nu_\alpha$  and  $\nu_\beta$  correspond to  $\text{Mn}^{2+}$  ions in positions *c* and *b*, respectively.

As a result we are left with one free parameter, since  $\Delta_\alpha$  and  $\Delta_\beta$  are connected by the relation  $\Delta_\alpha/\nu_\alpha = \Delta_\beta/\nu_\beta$ . It is more convenient in experiment to determine  $\Delta_{\alpha\beta}$  from the asymptotic form in weak fields and then calculate  $\Delta_\alpha$  and  $\Delta_\beta$ . The correctness of this procedure was verified by a computer calculation using Eq. (3); the results of this calculation fit best the experimental data at  $\Delta_\alpha^2 = 11.1/T$  kOe<sup>2</sup>/K,  $\Delta_\beta^2 = 10.0/T$  kOe<sup>2</sup>/K (solid curves in Fig. 1). The average coupling constant  $\Delta_{\alpha\beta}^2 = 10.5/T$  kOe<sup>2</sup>/K agrees satisfactorily with the value of the hyperfine gap in the AFMR spectrum.<sup>4</sup>

### SPIN-LATTICE RELAXATION OF Mn NUCLEI

Two NMR methods were developed for the measurement of spin-lattice relaxation. The first is to study the rate of restoration of the NMR signal after application of a high-power pulse, and the second is to measure the temporal characteristics of stimulated (three-pulse) echo. In substances with dynamic frequency shift of the NMR frequency measurements were previously made using both methods: NMR saturation<sup>7,9</sup> and stimulated echo,<sup>8</sup> but the results for one and the same sample were not compared. Such a comparison is necessary since, strictly speaking, different relaxations are being studied: relaxation of the average nuclear magnetization in the first case and relaxation of nuclear spin waves with small wave vectors in the second. In addition, the non-linear phenomena that accompany the formation of the stimulated echo can also contribute to the experimental results.

Using a high intensity of the spin-echo signal in  $\text{CsMnCl}_3$ , we made such a comparison. We observed the stimulated echo by the procedure described in Ref. 8. The intensity of the echo signal decreases exponentially with increasing delay between the second and third exciting microwave pulses, and the argument of the exponential yields the spin-lattice relaxation time  $T_1$ .

The procedure of measuring  $T_1$  using NMR saturation was based in our case on the dependence of the NMR frequency on the effective spin temperature. The irradiating microwave pulse of frequency  $f$  ( $\nu_\beta > f > \nu_1(H, T)$ ) overheats the Mn nuclei to a temperature  $T^*$  determined by the condition  $\nu_1(H, T^*) = f$ . The spin system then cools off and the NMR frequency drops again to the equilibrium value. If the spectrometer is tuned to an intermediate frequency  $f'$  ( $f > f' > \nu_1(H, T)$ ), it will record the NMR signal at a certain definite instant of time  $\tau$  after the end of the exciting pulse. The time of appearance of the echo signal at the frequency  $f'$  is determined by the formula

$$[\nu_1^{-2}(H, T) - (f')^{-2}] / [\nu_1^{-2}(H, T) - f^{-2}] = \exp(-\tau/T_1). \quad (6)$$

The experimental procedure was the following. The magnetic field [and hence  $\nu_1(H, T)$ ] and the frequency at which the echo  $f'$  was observed were fixed. For each frequency  $f$  we determined the position of the maximum of the

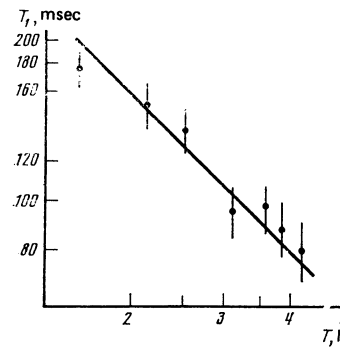


FIG. 2. Temperature dependences of nuclear spin-lattice relaxation time  $T_1$  at an NMR frequency 530 MHz.

intensity as a function of the delay  $\tau$  relative to the irradiating pulse. To check on the validity of Eq. (6) and to improve the accuracy, the measurements were made at several frequencies.

A comparison of the two procedures of measuring spin-lattice relaxation has shown that NSW with small  $k$  relax twice as fast as the average nuclear magnetization. In addition, the time  $T_1$  measured by a stimulated echo was found to increase with increasing power of the exciting microwave pulses. We have therefore used subsequently only the NMR saturation technique which, while more laborious, gives results that are more reliable and are easier to interpret.

The temperature and frequency dependences of the spin-lattice relaxation time  $T_1$  are shown in Figs. 2 and 3. Attention is called to the large values  $T_1 \approx 60$ – $80$  msec at 4.2 K, which exceed somewhat the record for the NMR frequency of the class of dynamic compounds with dynamic NMR frequency shift.<sup>7</sup> At the same time the relatively weak temperature dependence ( $T_1 \propto T^{-1}$ ) and frequency dependence of  $T_1$  are typical of extrinsic relaxation processes.<sup>1</sup> A similar picture was noted in  $\text{CSMnCl}_3$  investigations by electron-electron parallel pumping, where a large contribution of extrinsic relaxation processes was also observed.<sup>4</sup> All this indicates that by improving the quality of the  $\text{CsMnCl}_3$  samples it is possible to obtain a substance with record high relaxation characteristics.

Attention should also be called to the step near  $\nu^* = 480$  MHz on the plot of  $T_1$  vs frequency (Fig. 3). Although its size is of the order of our experimental error, nonetheless the fact that the results recur systematically indicates the presence of some singularity in the spin-lattice relaxation. Since an anomalous behavior of the spin-spin relaxation was observed in the same frequency region, we shall return to this question in the last section.

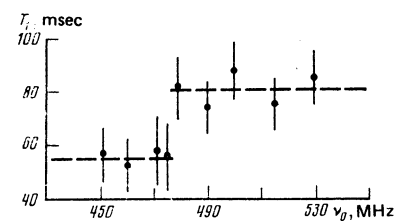


FIG. 3. Frequency dependence of  $T_1$  at  $T = 4.2$  K.

## SPIN-SPIN RELAXATION OF Mn<sup>55</sup> NUCLEI

The spin-spin relaxation in the Mn nuclei in CsMnCl<sub>3</sub> was determined at the NMR frequency from the decrease of the parametric spin echo signal (see Ref. 11 for details on the parametric-echo mechanisms and for the technique of its observation), and was determined in the region of the NSW from the threshold of their parametric excitation.

The spin-spin relaxation times  $T_2$  measured by the first method turned out to be quite long ( $T_2 \approx 15\text{--}50 \mu\text{sec}$  at  $T = 42 \text{ K}$ ). Their temperature dependences,  $T_2 \propto T^{-1.1 \pm 0.2}$ , agree with the data obtained with other substances with dynamic shift of the NMR frequency.<sup>8</sup> The frequency dependences of  $T_2$  are shown in Fig. 4, and it is easily seen that a resonant singularity is superimposed on the usual, in this case, monotonic growth  $T_2 \propto \delta\nu_0^{-1}$  (solid curves;  $\delta\nu_0$  is the NMR line width) near  $\nu_0 = 480 \text{ MHz}$ . The relative decrease of  $T_2$  and the position of the minimum do not depend on temperature in this case, and the width of the minimum decreases noticeably with decreasing temperature. No singularities whatever were observed in the position or width of the NMR line in the region  $\nu^* = 480 \text{ MHz}$ .

## RELAXATION OF NUCLEAR SPIN WAVES

The NSW were parametrically excited by the method of parallel microwave pumping in the frequency region  $\nu_k = \nu_p/2 = 400\text{--}520 \text{ MHz}$  and at temperatures 1.6–4.2 K. The signal from the microwave generator passed through a calibrated attenuator into a half-wave helical resonator with loaded  $Q \sim 500$ . When the magnetic field at the sample reached the threshold value  $h_c$ , parametric excitation of NSW was started. From the value of  $h_c$  we calculated the NSW relaxation rate ( $\Gamma$ ), using the formula

$$\Gamma = (h_c H / 2 \Delta_{\alpha\beta}) (\nu_{\alpha\beta}^2 - \nu_k^2)^2 / \nu_{\alpha\beta}^2 \nu_k. \quad (7)$$

The NMR spectrum of the investigated crystal is rather complicated in form (see above), but in the frequency region  $\nu_k < 520 \text{ MHz}$  it can be approximately described by Eq. (4). In this case the error in the relaxation rate  $\Gamma$ , due to the

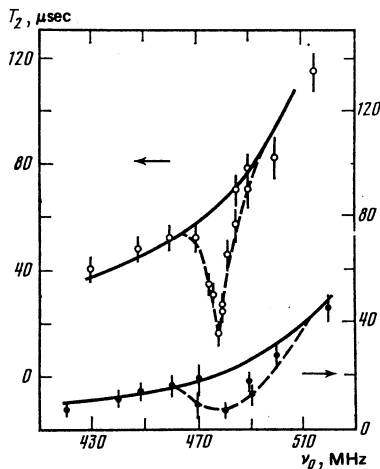


FIG. 4. Frequency dependence for the nuclear spin-spin relaxation time  $T_2$ : ● —  $T = 4.2 \text{ K}$ , ○ —  $T = 1.6 \text{ K}$ .

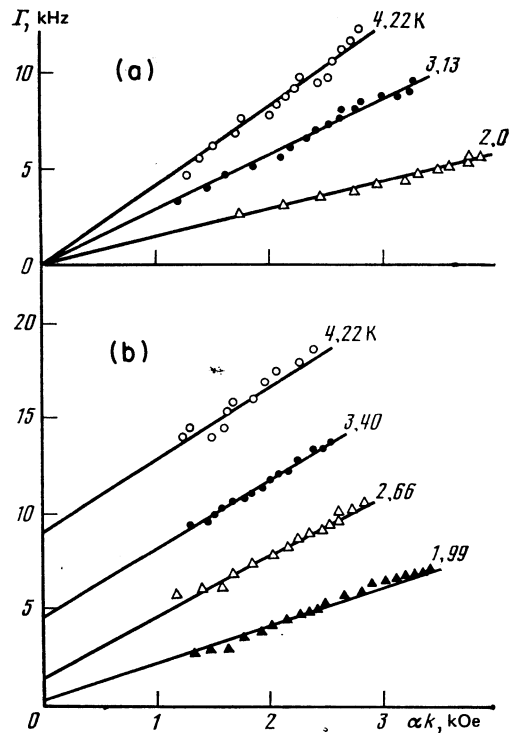


FIG. 5. Dependences of NSW relaxation rates on the wave number at various frequencies: a— $\nu_k = 504 \text{ MHz}$ ; b— $\nu_k = 475 \text{ MHz}$ .

deviation from the true spectrum, does not exceed its experimental error (see below).

The threshold field  $h_c$  was recorded by the modulation technique described in Ref. 13. To this end, an additional modulation field of frequency 100 kHz was applied to the sample. The modulation amplitude was chosen to be small enough ( $< 0.1 \text{ Oe}$ ) to make its influence on the threshold negligibly small. We estimate the relative accuracy of  $\Gamma$  at  $\pm 5\%$ , whereas the absolute accuracy was  $\pm 25\%$ .

Figure 5 shows plots of the NSW relaxation rates vs the wave number, obtained for two pump frequencies. The behavior of relaxation shown in Fig. 5a is satisfactorily described by the relation

CsMnCl<sub>3</sub>

$$\Gamma_1 = 1.2 \alpha k T^{1.2 \pm 0.2}, \quad (8)$$

where  $\Gamma_1$  is in kHz,  $\alpha k$  in kOe, and  $T$  in °K. Such a behavior of the relaxation was observed for NSW with frequencies  $\nu_k > \nu^* = 482 \text{ MHz}$ .

At lower frequencies ( $\nu_k < \nu^*$ ) the behavior of the relaxation becomes more complicated (Fig. 5b). One more process  $\Gamma_2$  is added to the relaxation  $\Gamma_1$ ; it is independent, within the limit of experimental error, of  $k$  and increases with temperature like  $T^{2.8 \pm 0.3}$  (Fig. 6). The frequency dependence of  $\Gamma_2$  is shown in Fig. 7. It can be seen that this mechanism is turned on abruptly at the frequency  $\nu^*$ , after which it grows and reaches at  $\nu_k = 430 \text{ MHz}$  a value  $\Gamma_2[\text{kHz}] = 0.22 T^3 [\text{K}]$ . Such a behavior of  $\Gamma_2$  gives grounds for assuming that at  $\nu_k > \nu^*$  this relaxation process is forbidden by the energy conservation law. This question is discussed in greater detail in the next section.

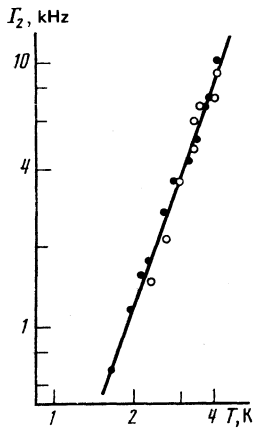


FIG. 6. Temperature dependence of the relaxation process  $\Gamma_2$ : ● —  $\nu_k = 448$  MHz; ○ —  $\nu_k = 475$  MHz.

It follows from the NSW relaxation theory developed in Refs. 14 and 15 that the main contribution to the relaxation at low temperatures and at  $k > 10^5$  cm $^{-1}$  should be made by elastic scattering of the NSW by localized nuclear spins. The NSW relaxation rate corresponding to this process is<sup>14</sup>

$$\Gamma = \frac{3}{8\pi} \frac{Am_B\gamma_n}{\gamma_n H_E} \frac{k_B}{\hbar} akT, \quad (9)$$

where  $k_B$  is the Boltzmann constant and  $a$  is the lattice constant. Using the values  $a = 5.2$  Å and  $A = \nu_{\alpha\beta}/\gamma_n M$  calculated in the cubic approximation, where  $M$  is the magnetic moment of the Mn $^{2+}$  ion, as well as other parameters of CsMnCl $_3$ , we obtain

$$\Gamma = 0.015kT, \quad (10)$$

where  $k$  is in cm $^{-1}$ ,  $T$  is in °K, and  $\Gamma$  is in kHz. It can be seen that the theoretical dependence of the relaxation on the temperature and on the wave number agrees satisfactorily with the experimental (8).

For a quantitative comparison we must know the value of the exchange constant  $\alpha$ , which has not been measured hitherto in CsMnCl $_3$ . It can be estimated by using the results of Ref. 4, where phonon emission by parametric magnons is observed. This process is allowed by the conservation laws if the magnon group velocity  $v(k)$  exceeds the velocity  $u_{\min}$  of the slowest sound, i.e.,

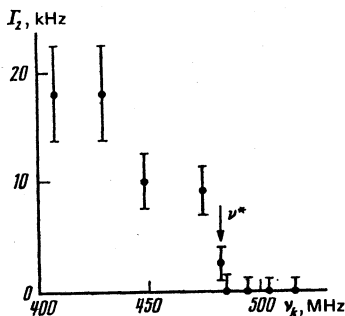


FIG. 7. Frequency dependence of the relaxation process  $\Gamma_2$  at 4.2 K.

TABLE I. Longitudinal and transverse sound velocities in CsMnCl $_3$  at room temperatures ( $k$  is the wave propagation vector,  $e$  is the polarization vector).

Propagation direction and polarization		$u$ , 10 $^5$ cm/sec
Transverse sound	$k \perp C_3, e \perp C_3$	$1.90 \pm 0.05$
	$k \perp C_3, e \parallel C_3$	$1.74 \pm 0.05$
	$k \parallel C_3, e \perp C_3$	$1.75 \pm 0.1$
Longitudinal sound	$k \parallel C_3$	$3.6 \pm 0.15$
	$k \perp C_3$	$3.0 \pm 0.15$

$$v(k) = (2\pi\alpha\gamma^2/\Omega_k) \alpha k \geq u_{\min}, \quad (11)$$

where  $\Omega_k$  is the frequency of the parametric magnons. The value of  $\alpha k_c$  at which the velocities  $v$  and  $u_{\min}$  become equal was obtained in Ref. 4.

To determine the value of  $\alpha$  from (11) we measured the speed of sound in CsMnCl $_3$  at room temperatures. The longitudinal-sound measurements were made at 3 MHz, and the transverse at 35 MHz. The results of these measurements are gathered in Table I. For the lowest sound speed  $u_{\min} = 1.74 \times 10^5$  cm/sec we obtain from (11)  $\alpha = 1.25 \times 10^{-5}$  kOe cm. Substituting this value in (8) we obtain for the numerical coefficient a value 0.015 which coincides with the theoretical value, see (10).

We note that another frequency dependence,  $\Gamma_1 \propto \nu_k^3$ , was predicted for the process considered in Ref. 15. In our experiment, however,  $\Gamma_1$  remained constant (within  $\pm 15\%$ ) in the entire frequency range, whereas the theory predicts a change of  $\Gamma_1$  by more than two times.

#### POSSIBLE MECHANISMS OF ANOMALOUS BEHAVIOR OF RELAXATION NEAR $\nu^*$

The experimental results described above offer evidence that the NSW relaxation, as well as the spin-spin and spin-lattice relaxation times, have significant anomalies near the frequency  $\nu^* \approx 4.82 \pm 3$  MHz. The singularities in  $\Gamma$  and  $T_1$  have a steplike character with increasing  $\Gamma$  (with decreasing  $T_1$ ) at frequencies  $\nu < \nu^*$ , while  $T_2$  has a local minimum near  $\nu^*$ . We consider below two mechanisms that make it possible to explain these results qualitatively.

If it is assumed that excitations with characteristic frequency 70 MHz are present in our crystal, then the process of their coalescence into NSW is possible only at frequencies  $\nu < \nu^*$ , while at higher frequencies it is forbidden by the energy conservation law. Such excitations might be transitions between the hyperfine structure of the neutral Mn atoms, which takes place at a frequency 72 MHz (Ref. 16). Therefore the presence of an admixture of Mn neutral atoms in the CsMnCl $_3$  might explain the steplike behavior of  $\Gamma$  and  $T_1$  near  $\nu^*$ .

The character of the anomaly in the behavior of  $T_2$  is qualitatively similar to the known cross relaxation of NMR in diamagnets. It can therefore be assumed this singularity to be due to interaction with some resonant excitation at the frequency  $\nu^*$ . This frequency can be possessed by NMR in the Mn $^{3+}$  ion, the hyperfine field for which is 20–30% lower than for the Mn $^{2+}$  ion. The steplike character of the singu-

larities of  $T_2$  and  $T_1$  can then be due to the presence of a dynamic shift of the NMR frequency of the  $Mn^{3+}$  admixture. So far, however, there are no theoretical or experimental indications of such a behavior in other substances.

Thus, the contamination of  $CsMnCl_3$  by neutral Mn atoms or by  $Mn^{3+}$  ions might explain qualitatively the singularities in the relaxation of the nuclear system near  $\nu^*$ . The final answer to this question, however, calls for numerical calculations.

The authors are most grateful to B. V. Beznosikov for supplying the  $CsMnCl_3$  samples as well as to V. I. Ozhogin and V. L. Safonov for helpful discussions.

<sup>11</sup>Typical relations for intrinsic relaxation processes are  $T_1 \propto T^{-(4-5)}$  and  $T_1 \propto H^{-(1-2)}$  (see Refs. 7 and 10).

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Translated by J. G. Adashko