Spin wave relaxation in antiferromagnetic $CsMnF_3$

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Parametric excitation of electron and electron-nuclear spin waves and premature saturation of the principal resonance were investigated in the hexagonal antiferromagnet $CsMnF_3$ with easy-plane anisotropy. The experiments were performed at temperatures 1.2 to 2.2°K in the frequency range ν_p = 9 – 50 GHz. It was observed in the experiments on the parametric excitation of the electron spin waves that the absorption sets in and vanishes in a jumpwise, "rigid" manner, apparently owing to the presence of a damping mechanism that is turned off at a definite amplitude of the parametrically excited spin waves. The dependences of the turned-off part $\Delta\nu_1$ of the relaxation and of the stationary part $\Delta\nu_2$ on the temperature and on the static magnetic field are investigated. There is no "rigidity" effect in the parametric excitation of electron-nuclear paris of spin waves and in antiferromagnetic-resonance saturation. We discuss the causes of this phenomenon and analyze the relaxation frequencies $\Delta\nu$ obtained in these experiments. The exchange constant in the spin-wave spectrum is determined from the points of intersection of the magon and phonon spectra and from the known speed of sound, and is found to be $\alpha_\perp = 0.95 \times 10^{-2}$ Oe-cm ± 5%.

INTRODUCTION

One of the central problems in the study of the magnetic properties of antiferromagnetic dielectrics is the determination of the nature of the relaxation in a spin-wave system. We investigated theoretically various forms of interaction within this system—three-magnon and four-magnon processes, and also interactions with participation of phonons, scattering by impurities, etc. The theory enables us to obtain for each of these processes the dependence of the spin-wave lifetime τ or of the relaxation frequency $\Delta\nu_k = 1/2\pi\tau$ on the temperature, on the static magnetic field, on the wave vector of the spin wave, and on different intrinsic parameters of the substance, such as the exchange field, the anisotropy field, etc. Thus, experimental investigation of the relaxation frequency in antiferromagnets is of great interest.

The methods most widely used at present for the study of relaxation in antiferromagnets are: 1) determination of the antiferromagnetic resonance (AFMR) line width $\Delta\nu_0$, 2) determination of the threshold field h_c^{ee} of the parametric excitation of the electron spin waves, 3) determination of the threshold field h_c^{ne} of the parametric excitation of electron-nuclear spin waves, and 4) determination of the threshold field h_c^S of the "premature saturation" of the AFMR.

1. The first method does not seem promising to us because from the width of the AFMR line we can calculate only the relaxation frequency of the homogeneous precession and the magnetostatic modes. In addition, as shown by Kotthaus and Jaccarino [1], the line widths observed in experiments on AFMR are usually determined not by the true processes of relaxation in antiferromagnets, but by the inhomogeneous broadening and by the connection of the homogeneous precession with the degenerate magnetostatic modes.

2. The most promising method is, in our opinion, the second one. It is based on the dependence of the threshold microwave field h_c^{ee} at which parametric excitation of the electron spin waves with wave vector **k** sets in on the relaxation frequency $\Delta\nu_k^e$. Using the parallel-pumping method [2,3] in the microwave band, we can excite the spin waves in a broad wave-vector interval, up to 10^6 cm^{-1}.

3. In antiferromagnets in which the magnetic ions have a strong hyperfine interaction (for example, Mn^{++}), the nuclear-spin system is also ordered. In such antiferromagnets it is possible to produce simultaneously, by the parallel-pumping method, parametric excitation of electron and nuclear spin waves. The threshold field of this process is $h_c^{ne} \propto (\Delta\nu_k^e \cdot \Delta\nu_k^n)^{1/2}$. This makes it possible to determine the value of $\Delta\nu_k^e$ if $\Delta\nu_k^n$ is known from other experiments

4. As shown by Heeger [4], starting with a certain microwave field amplitude h_c^S, the imaginary part of the hf susceptibility at resonance χ'' begins to decrease with increasing h. According to Suhl [5] this process is due to the conversion of two magnons with $\mathbf{k} = 0$ into a pair of magnons that are degenerate with respect to them, with wave vectors **k** and **−k**. The threshold field h_c^S of this process is determined by the frequencies $\Delta\nu_0$ and $\Delta\nu_k$ of the homogeneous-precession relaxation and of the degenerate spin waves, respectively. At the present time it is not quite clear how to determine the quantity $\Delta\nu_0$ in the expression for h_c^S, so that $\Delta\nu_k$ is not quite uniquely determined by this method.

The experiments were performed by us with hexagonal $CsMnF_3$ which is presently among the most thoroughly investigated antiferromagnets. The absence of orbital effects, owing to the S state of the Mn^{++} ion, the uniaxial easy-plane anisotropy that causes the gap in the low-frequency branch of the AFMR spectrum to be determined only by the hyperfine interaction, and the absence of a Dzyaloshinskiĭ field, all simplify the theoretical analysis of the thermodynamic properties of $CsMnF_3$ appreciably and make it an almost ideal object for the study of spin-wave relaxation.

The spin-wave spectrum in $CsMnF_3$ consists of six branches. In our experiments we excited spin waves of the low-frequency branch of the spectrum, the frequency of which is determined by the formula

$$(\nu_1/\gamma)^2 = H^2 + H_\Delta^2 + \alpha^2 k^2, \quad (1)$$

where $H_\Delta^2 = 6.4\ T^{-1}$ [kOe^2] is the gap due to the hyperfine interaction [6], α is the exchange constant, and $\mathbf{k}$ is the spin-wave vector.

For antiferromagnets of the $CsMnF_3$ type, the expressions connecting the threshold fields of the processes described above with the spin-wave relaxation frequency take the form

$$h_c^{ee} = \frac{\nu_p \Delta\nu_k^e}{2\gamma^2 H} \quad [7], \tag{2}$$

$$h_c^{ne} = 2\frac{(\Delta\nu_k^n \Delta\nu_k^e)^{1/2}}{\gamma}\left(\frac{\nu_k^n \nu_k^e}{\gamma^2 H_\Delta^2}\right)^{1/2}\frac{(\nu_k^e)^2}{\gamma H \nu_0^n} \quad [8], \tag{3}$$

$$h_c^S = 4\Delta H_0\left(\frac{\gamma \Delta H_k}{\nu_0}\right)^{1/2} \quad [4], \tag{4}$$

where ν_p is the pump frequency. The purpose of the present study was to investigate the kinetics of the excitation of parametric spin waves and to determine the relaxation frequencies of the electron spin waves from the threshold fields h_c^{ee}, h_c^{ne}, and h_c^S.

PROCEDURE

The experiments were performed with direct-amplification spectrometers for the 3-cm and 8-mm bands.

The parametric excitation of the electronic spin waves was observed with the setup described in detail earlier [9]. A $CsMnF_3$ single crystal was secured with BF-4 adhesive to the bottom of a cylindrical resonator with $Q \sim 10\,000$ in the antinode of the magnetic field of the H_{012} mode in such a way that the fields h and H were in the basal plane of the crystal. The microwave source was a klystron oscillator operating in the millisecond-pulse regime. The pump frequency ν_p was varied in the interval from 25 to 50 GH_Z. The microwave pulse passing through the resonator was detected and fed to an oscilloscope. Absorption of microwave power by the sample, corresponding to parametric excitation of spin waves, was revealed on the oscilloscope by the appearance of a sharp decrease of the pulse amplitude. The dependence of the power absorbed by the sample on the static magnetic field H was registered in a number of experiments with an automatic x-y recorder. The instantaneous value of the amplitude h of the microwave field at the sample was determined by the method described in [9]. The absolute accuracy with which the field h was measured at the sample was ~15%, whereas the relative change of h in the series of experiments was measured with accuracy ~5%.

Parametric excitation of electron-nuclear spin waves at $h \parallel H$ was observed with a setup analogous to that described above. The pump frequency ν_p was ~9.3 GHz. Rectangular microwave pulses of duration $\tau \sim 100$ μsec were applied to the resonator with the sample. The use of rectangular pulses has made it possible to employ the usual procedure for the measurement of microwave power supplied to the resonator—we measured the average power at the resonator input and the off-duty factor of the pulses. The accuracy with which the microwave field h at the sample was determined was ~15%.

Premature saturation of AFMR was observed at a frequency 36 GHz, the fields $\mathbf{h}$ and $\mathbf{H}$ being perpendicular and both lying in the basal plane of the crystal. We used the same resonator as for the observation of the excitation of electron spin waves. The microwave generator was swept near the resonant frequency. The rate of deviation of the generator frequency was high enough, $\sim 10^3$ GHz/sec, to prevent overheating the sample, and the time required to scan the resonator resonance curves was ~3 μsec. The repetition frequency was ~100 Hz. The detected signal, which was proportional to the power P_1 passing through the resonator, was fed to an oscilloscope. The oscilloscope screen displayed the frequency characteristic $P_1(\nu)$ of the resonator. When a static magnetic field H_0 corresponding to the AFMR was applied to the sample, the resonator Q decreased, and this led to a decrease in the amplitude P_{1max} of the $P_1(\nu)$ curve. In the experiment, we measured dependence of the ratio $P_{1max}(H = H_0)/P_{1max}(0)$ on the microwave power fed to the resonator. The threshold value of the field h_S^S, at which premature saturation of the resonance took place, was determined from the break on the plot of this dependence, with accuracy ~2 dB.

The overheating of the sample was monitored against the change of H_0 and the known $H_0(T)$ relation, and did not exceed 0.2°K. To improve the heat transfer from the sample, the resonators were filled with superfluid helium in all the experiments. The experiments were performed in the temperature interval 1.2–2.1°K. The temperature was determined from the helium saturated-vapor pressure accurate to ~0.05°K. The static magnetic field was produced by a laboratory electromagnet and was measured accurate to ~1%.

EXPERIMENTAL RESULTS

1. Parametric excitation of electron spin waves was observed in the range 25–50 GHz. In these experiments we observed that spin-wave excitation, just as in $MnCO_3$ [10], is "hard" in character. This is manifest in the jumplike (within a time ~20 μsec) onset and vanishing of the absorption in the sample at different critical values of the microwave field on the sample, h_{c1}^{ee} and h_{c2}^{ee} respectively. The oscillograms of the pulses at the input and output of the resonator were analogous to those shown in Figs. 1 and 2 of [10].

Figure 1 shows the relation between the field h on the sample and the time τ_1 from the start of the pulse to its drop-off corresponding to the onset of absorption in the sample. The investigations were carried out at different temperatures and at different values of

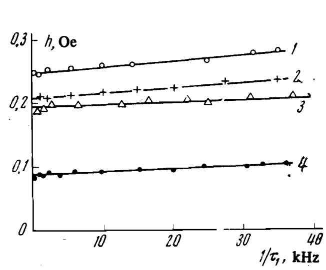

FIG. 1

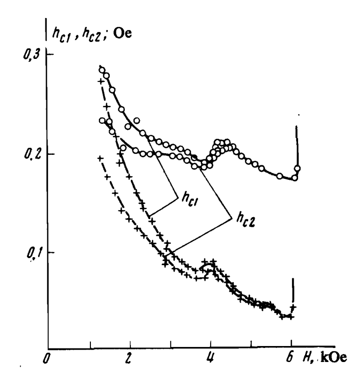

FIG. 2

FIG. 1. Reciprocal of the time from the start of the pulse to its drop-off vs. the microwave filed on the sample: 1–T = 1.55°K, H = 1.97 kOe; 2–T = 1.55°K, H = 4.44 kOe; 3–T = 1.20°K, H = 1.97 kOe; 4–T = 1.20 K, H = 4.44 kOe.

FIG. 2. Plots of the critical fields h_{c1}^{ee} and h_{c2}^{ee} against the static field at two temperatures: ○–T = 1.7°K, + –T = 1.25°K.

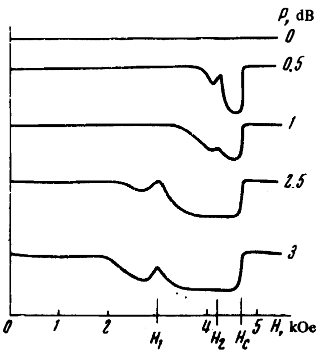

FIG. 3

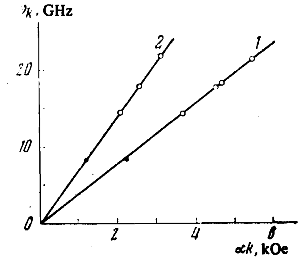

FIG. 4

FIG. 3. Automatic-recorder plots of above-threshold absorption at different microwave power levels; $T = 1.5°K$, $\nu_p = 28.8$ GHz.

FIG. 4. Dispersion law $\nu_k(\alpha k)$ for the anomalies at the fields H_1 and H_2; ●—data of [11].

the static field. The results show that h and $1/\tau_1$ are linearly related.

Figure 2 shows plots of h_{c1}^{ee} and h_{c2}^{ee} against the static field H for two temperatures, at a pump frequency $\nu_p = 36.2$ GHz. At $T = 1.25°K$ in fields of 4 and 5.5 kOe one can see anomalies of resonant character on the $h_{c1}^{ee}(H)$ and $h_{c2}^{ee}(H)$ plots. These anomalies become more clearly manifest in plots of the power absorbed by the sample against the static field. Figure 3 shows the corresponding plots obtained with an x-y recorder at different excesses above the threshold power. The absorption amplitude has a minimum at the fields H_1 and H_2, in accord with the increase of the threshold field h_{c1}^{ee} at these points.

To explain the nature of the indicated peaks, we performed experiments at different pump frequencies ν_p in the range from 25 to 50 GHz. The experimental results are shown in Fig. 4, where the fields H_1 and H_2 are converted into the quantity αk in accordance with formula (1). The same figure shows the data by Seavey [11].

2. In experiments in which electron and nuclear spin waves were observed simultaneously, we did not see any symptoms of "hard" excitation. The process is characterized by a single threshold field h_c^{ne}. Figure 5 shows plots of the field h_e^{ne} against the static field H for different temperatures.

3. Figure 6 shows the plots of the imaginary part of the susceptibility χ'' in AFMR against the microwave power fed to the resonator at $T = 1.2$ and $1.9°K$. Starting with a certain power value P_c^S, the value of χ'' begins to decrease with increasing P. We interpret the field h_c^S on the sample, corresponding to the power P_c^S, as the threshold field for the premature saturation of AFMR. The values of the threshold fields were found to be

$$h_c^s(1.2°\,K) = 0.07\,Oe, \quad h_c^s(1.9°\,K) = 0.2\,Oe.$$

DISCUSSION OF RESULTS

1. Since the excitation of electron spin waves is "hard" in character, the results of the experiments will be considered under the assumption that the spin-wave relaxation consists of two parts:

$$\Delta\nu^e = \Delta\nu_1^e + \Delta\nu_2^e, \quad (5)$$

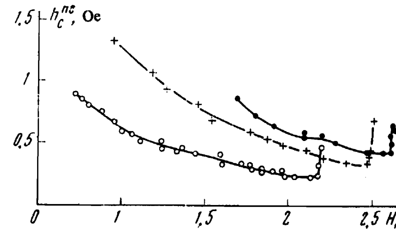

FIG. 5. Critical field h_c^{ne} against the static magnetic field at different temperatures: ●—1.25°K, ○—1.55°K, + —1.7°K.

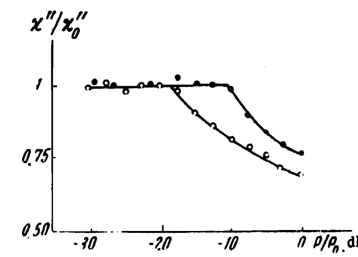

FIG. 6. Plot of the imaginary part of the dynamic susceptibility χ'' against the microwave power ($P_0 = 24$ mW): ●—$T = 1.9°K$, ○—$T = 1.2°K$.

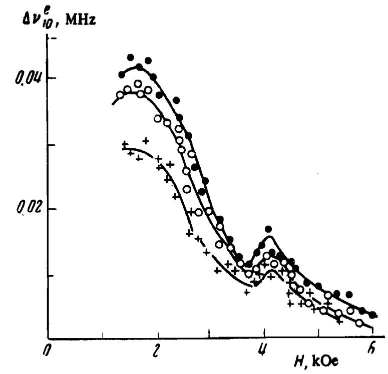

FIG. 7. Dependence of the relaxation $\Delta\nu_{10}^e$ on the static magnetic field at different temperatures: ●—1.25°K, ○—1.55°K, + —1.7°K.

where $\Delta\nu_2^e$ depends little on the number n_k of excited spin waves, while $\Delta\nu_1^e$ decreases with increasing n_k, from an initial value $\Delta\nu_{10}^e$ corresponding to $n_k = 0$ to zero. This assumption was discussed in detail in [10]. The question of the nonlinear negative damping in ferromagnetic systems was considered by a number of workers [12,13]. From the results of our experiments we can calculate the relaxation frequencies $\Delta\nu_{10}^e$ and $\Delta\nu_2^e$ from the formulas

$$\Delta\nu_2^e = 2h_{c2}^{ee}H/\gamma^2\nu_p,$$
$$\Delta\nu_{10}^e = 2h_{c1}^{ee}H/\gamma^2\nu_p - \Delta\nu_2^e. \quad (6)$$

It is not quite correct to determine the relaxation frequency $\Delta\nu_2^e$ from the value of the critical field h_{c2}^{ee}, and more accurate results were obtained in [10] by extrapolating the power absorbed by the sample to zero. However, as shown in the same reference, the error due to such a simplification is small.

Figures 7 and 8 show the calculated values of $\Delta\nu_{10}^e$ and $\Delta\nu_2^e$ as functions of the static magnetic field. The relaxation frequency $\Delta\nu_{10}^e$ decreases with increasing magnetic field and vanishes at $H = H_c$, corresponding to $k = 0$. The value of $\Delta\nu_{10}^e$ also decreases with increasing temperature.

The relaxation frequency $\Delta\nu_2^e$ depends linearly on the square of the static magnetic field up to fields ~4 kOe, and can be represented in the form

$$\Delta\nu_2^e = \Delta\nu_{20}^e(T) + \beta(T)H^2. \quad (7)$$

The value of $\Delta\nu_{20}^e$ varies little with temperature and is equal to ~0.1 MHz. This relaxation frequency corre-

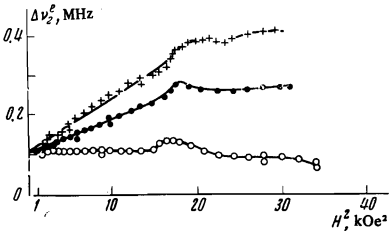

FIG. 8. Dependence of the relaxation $\Delta\nu_2^e$ on the square of the static field H^2 at different temperatures: ○–1.25°K, ●–1.55°K, +–1.70°K.

sponds to a magnon mean free path $\lambda \sim 2$ mm at $H = 0$ ($k \sim 6\times 10^5$ cm^{-1}), which is close to the sample dimensions. It can therefore be concluded that $\Delta\nu_{20}^e$ is determined in the main by the scattering of the magnons from the crystal boundaries. In the investigated temperature interval 1.2–2.2°K, β is proportional to $T^{6.7}$.

We note that the described results not only agree qualitatively with the results obtained earlier with $MnCO_3$ [9,10], but are also close to them quantitatively. This agreement, in spite of the appreciable difference between the crystallographic structures of these substances, seems to point to a common nature of electron spin wave relaxation. In the described experiments, at $\nu_p = 36$ GHz, the field h_c^{ee} was measured for spin waves in the k interval from 0.5×10^5 to 6×10^5 cm^{-1}. The upper limit was calculated from formula (1) for a field $H \sim 1$ kOe (in weaker fields, the samples may have more than one domain [6]). The lower limit is determined by the region of degeneracy as $k \to 0$ [14,15], the presence of which leads to an ambiguity in the determination of k between 0 and 0.5×10^5 cm^{-1}.

The conditions under which the relaxation was measured, namely $h\nu_k \sim k_B T$, $H \neq 0$, and $k \neq 0$, do not reduce to the limiting cases considered by the theory at the present time [16–18], so that no conclusion can be drawn as yet concerning the nature of the relaxation.

2. The increase of the threshold field h_{c1}^{ee} at the fields H_1 and H_2 corresponds to an increase in the relaxation of the spin waves at these points. The linear $\nu_k(\alpha k)$ relation corresponding to these anomalies allows us to state that we are observing an intersection of the magnon and phonon spectra.

To determine the type of phonons connected with the spin waves excited in our experiments, we write down the general expression for the Hamiltonian of the magnetoelastic interaction determined by the crystal symmetry (symmetry space group D_{6h}^4):

$$\begin{aligned} H_{me} = {} & u_{zz}[a_1 l_{1z}^2 + a_2 l_{2z}^2 + a_3(l_{1x}^2 + l_{1y}^2) \\ & + a_4(l_{2x}^2 + l_{2y}^2) + a_5(l_{1x}l_{2x} + l_{1y}l_{2y}) + a_6 l_{1z}l_{2z}] + (u_{xx} + u_{yy})[b_1 l_{1z}^2 + b_2 l_{2z}^2 \\ & + b_3(l_{1x}^2 + l_{1y}^2) + b_4(l_{2x}^2 + l_{2y}^2) + b_5(l_{1x}l_{2x} + l_{1y}l_{2y}) + \\ & + b_6 l_{1z}l_{2z}] + c_1(u_{yz}l_{1y}l_{1z} + u_{xz}l_{1x}l_{1z}) + c_2(u_{yz}l_{2y}l_{2z} + u_{xz}l_{2x}l_{2z}) + c_3(u_{yz}l_{2y}l_{1z} \\ & + u_{xz}l_{2x}l_{1z}) + c_4(u_{yz}l_{1y}l_{2z} + u_{xz}l_{1x}l_{2z}) + d_1[(u_{xx} - u_{yy})(l_{1x}^2 - l_{1y}^2) + 4u_{xy}l_{1x}l_{1y}] \\ & + d_2[(u_{xx} - u_{yy})(l_{2x}^2 - l_{2y}^2) + 4u_{xy}l_{2x}l_{2y}] + \\ & + d_3[(u_{xx} - u_{yy})(l_{1x}l_{2x} - l_{1y}l_{2y}) + 2u_{xy}(l_{1x}l_{2y} + l_{1y}l_{2x})]. \end{aligned} \quad (8)$$

The introduction of the two vectors $\mathbf{l}_1$ and $\mathbf{l}_2$ is due to the presence of two crystallographically nonequivalent positions of the Mn^{++} ion in the unit cell. Concrete expressions for $\mathbf{l}_1$ and $\mathbf{l}_2$ in terms of the magnetic moments of the individual ions are given in [19]. The expression for H_{me} is invariant to rotation about the axis $z \parallel C_5$. Using this fact, we choose the x-axis direction along the magnetic field H.

We represent, as usual, $\mathbf{l} = \mathbf{l}_1 + \mathbf{l}_2$ in the form $\mathbf{l} = \mathbf{l}_0 + \lambda\exp[i(\omega t - \mathbf{k}\cdot\mathbf{r})$. It is known that $\mathbf{l}_0 \perp \mathbf{H}$, and in spin waves corresponding to the low-frequency branch of the spectrum, only the component $\lambda_x \parallel H$ differs from zero, i.e., $\mathbf{l} = (\lambda_x\exp[i(\omega t - \mathbf{k}\cdot\mathbf{r})], l_{0y}, 0)$. Substituting the vector $\mathbf{l}$ expressed in this form in H_{me} and retaining only the terms proportional to the first power of $\exp[i(\omega t - \mathbf{k}\cdot\mathbf{r})]$, we can easily show the following: a) the constants d_1, d_2, and d_3 are responsible in our case for the interaction; b) the spin waves interact only with longitudinal and transverse phonons whose wave vector and polarization vector lie in the basal plane. Using the sound-velocity values v_s and v_l given in [11], we obtain for the value of the constant

$$\alpha_\perp = 0.95\cdot 10^{-2}\ \text{Oe}\cdot\text{cm} \pm 5\%.$$

3. When a pair of electron and nuclear spin waves are excited, the frequencies and the wave vectors are connected by the relations

$$\nu_p = \nu_k^e + \nu_k^n(T, H), \quad \mathbf{k}^e = \mathbf{k}^n,$$

where $\nu_k^n(T, H)$ is the frequency of the excited nuclear branch and is determined by the formula [20]

$$\nu_k^n(T, H) = \nu_{n0}\left(1 - \frac{\gamma^2 H_\Delta^2}{(\nu_k^e)^2}\right)^{1/2}, \quad \nu_{n0} = 666\ \text{MHz}. \quad (9)$$

It follows from the results given in Fig. 5 that: 1) the absorption produced in the sample takes place up to fields $h_c^{ne}(T)$ corresponding to excitation of spin waves with $k = 0$; 2) in the nuclear-spin system there exist spin waves up to $k = 3.5\times 10^5$ cm^{-1}; 3) the threshold field h_c^{ne} increases with increasing temperature and wave vector.

Analogous experiments at $\nu_p = 9.35$ GHz at two temperatures, 1.7 and 4.2°K, were performed by Seavey [2]. The value of h_c^{ne} obtained by us at $T = 1.7$°K is close to that given in [2]. In the same paper, Seavey determined the relaxation rate of the electron spin waves, and was therefore able to separate the quantity $\Delta\nu_k^n$ in accordance with formula (3). His result, however, exceeded by approximately 6 times the value of $\Delta\nu_k^n$ measured directly in [21].

In our opinion, the essential result of the experiments on the excitation of electron-nuclear spin waves is the absence of "hardness." The point is that the presence of a relaxation component $\Delta\nu_1^e$ that becomes turned off should lead to "hard" excitation of the electron-nuclear spin waves. It follows therefore either that the electron spin waves with frequency $\nu_k = \nu_p - \nu_n \approx 9$ GHz, which are excited simultaneously with the nuclear waves, no longer have a relaxation component that becomes turned off, or else that the above-threshold susceptibility is so low that the presence of this relaxation component does not lead to the appearance of a jump in the absorption.

4. From the data obtained in experiments on premature saturation of AFMR (Fig. 6) it follows that the threshold field h_c^S increases with increasing temperature. From the value of h_c^S it is possible to estimate, by means of formula (4), the value of $\Delta\nu_k^e$ for spin waves of frequency $\nu_k = 36$ GHz, into which the homogeneous precession decays. Assuming for ΔH_0 the experimentally observed value ~30 Oe, we obtain $\Delta\nu_k(1.2°K) \sim 10$ kHz.

This quantity is smaller by one order of magnitude than the $\Delta\nu_k^e$ obtained in experiments on parametric excitation. It appears that this is due to the inhomogeneous broadening of the AFMR line. A similar conclusion is also reached by other authors [4,22,23]. To obtain a value on the order of 0.1 MHz (the minimal observable relaxation) for $\Delta\nu_k^e(1.2°K)$ it is necessary to assume $\Delta H_0 \approx 10$ Oe.

"The hardness" effect is likewise not manifested in these experiments, owing to the fact that magnons with small **k** corresponding to the degeneracy region are produced upon saturation, and it follows from Fig. 7 that $\Delta\nu_1^e$ decreases as $\mathbf{k} \to 0$.

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