# SCATTERING OF PARAMETRIC SPIN WAVES BY NUCLEAR MOMENTS

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Interaction between nuclear magnetic moments and parametric spin waves in magnetically ordered crystals is observed for the first time by means of a novel electron-nuclear double resonance. It is found experimentally that the rate of transverse nuclear relaxation induced in the ferrimagnetic  $Y_3Fe_5O_{12}$  by parametric spin waves exceeds by three orders of magnitude the corresponding nuclear relaxation due to thermal magnons. It is demonstrated theoretically that the effect consists in two-magnon scattering of parametric spin waves by nuclear moments. The magnitude of  $1/T_2$  due to scattering of spin waves by nuclei is determined. Satisfactory agreement between the calculated and experimental values of  $1/T_2$  is obtained. Some new mechanisms of longitudinal nuclear relaxation involving parametric spin waves are proposed. The presence of a dynamic shift and broadening of the NMR lines induced by microwave pumping is demonstrated. The possibilities of investigating the dynamics of nuclear magnetization motion in the presence of radio-frequency pulses are discussed.

 $\mathbf{T}$  HE most important features of the behavior of the system of nuclear moments in magnetically-ordered substances are determined by the interaction between the nuclear spins and the magnetically-ordered electron system. In particular, the value of the local magnetic field at the nuclei, and also a large number of nuclearrelaxation processes depend on the electronic magnetization and on the fluctuations of the electron spins. Under ordinary conditions when  $\mathbf{T}\ll\mathbf{T_{c}}$  the state of the electronic system is described in terms of the spin waves (magnons). In this case many interactions of the nuclear moments with the magnetic electron system reduce essentially to the interaction of the nuclear moments with the magnons excited in the sample as a result of the thermal energy. There is an extensive literature devoted to an analysis of these effects<sup>[1-4]</sup>. However, spin waves can be excited in a sample artificially, for example by parametric pumping<sup>[5-7]</sup>. But how the nuclear spins interact with these artificially excited parametric spin waves and what information can be obtained by investigating this interaction is apparently still unknown, since until recently there have been no reports of any theoretical or experimental investigations of this question.

We reported in a brief note<sup>[8]</sup> the first observation of the influence of parametric spin waves on nuclear transverse relaxation in  $Y_3Fe_5O_{12}$ . In the present article we describe in detail the experimental procedure, give the experimental results, consider the theory of nuclear transverse relaxation due to scattering of parametric spin waves by nuclear moments, and discuss the possibilities of processes of nuclear longitudinal relaxation and the features of observation of nuclear echo in the presence of microwave pumping.

## 1. EXPERIMENTAL CONDITIONS

The apparatus for the investigation is a combination of two pulsed setups. One is a spectrometer for the observation of nuclear spin echo in the 50-80 MHz range and the other is a setup for parallel pumping of spin waves in ferrites. A block diagram of the complete apparatus is shown in Fig. 1. The setup for parallel pumping includes a pulsed microwave generator operating at 9640 MHz and delivering a power up to 1 kW in the pulse, and the necessary matching elements, attenuators, resonator, and electromagnet. The direction of the alternating microwave field coincides with the direction of the constant magnetic field  $H_0$  (parallel pumping). The threshold of excitation of the parametric spin waves is detected by observing the waveform of the microwave pulse on an oscilloscope screen.

The sample is placed in a rectangular resonator. A coil of three turns of copper wire is wound around the sample to excite the radio frequency field  $(H_{rf} \perp H_0)$  at the NMR frequency and to observe the nuclear- echo signal. The coil is connected through a cable to the nuclear- echo spectrometer. The main measurements were performed at ~ 140°K. This temperature was produced by blowing nitrogen vapor around the resonator. The sample was a sphere (3 mm diameter) of the polycrystalline ferrite  $Y_3Fe_5O_{12}$  enriched with the isotope Fe<sup>57</sup> to 87.1%. The nuclear echo was observed from the Fe<sup>57</sup> nuclei situated in the d-sublattice of the



FIG. 1. Block diagram of experimental setup. The programmed generator, modulator, radio-pulse generator, measuring receiver, and oscilloscope compose the nuclear-spin-echo spectrometer, and the rest of the equipment is the apparatus for excitation of parametric spin waves. yttrium iron garnet. The NMR frequency at  $T\approx 140^\circ$  amounts to 63–62 MHz.

The experiments consist of investigating the intensity of the nuclear-echo signal and measuring the times of the nuclear relaxation in the presence of pulsed excitation of parametric spin waves. The triggering of the microwave generator is suitably synchronized with the triggering of the rf pulses , and the position of the microwave pulse can be delayed by the required amount relative to the instant of triggering of the rf pulses. The duration of the microwave pulses was in most experiments 1  $\mu$  sec. The repetition frequency of the series of pulses was several Hz.

### 2. EXPERIMENTAL RESULTS

When a microwave pulse of power exceeding the threshold of the spin-wave instability is turned on, we observed a change (as a rule, a decrease) in the intensity of the nuclear-echo signal. The degree of decrease of the signal depends on the power of the microwave pulse and its position on the time scale relative to the rf pulses. There is no influence of the parallel pumping if the latter is applied prior to the first rf pulse or after the echo signal. This circumstance, in particular, indicates that the heating of the sample under the influence of the microwave power has no effect. The dependence of the signal-echo intensity in relative units (relative to the intensity of the echo without pumping) on the position of the microwave pulse  $(T_p)$  on the time scale is shown in Fig. 2. In this case we observe the usual echo formed after application of 90 and  $180^{\circ}$  pulses (the first and second pulses are 4 and 8  $\mu$  sec long).

As seen from the figure, the intensity of the echo decreases smoothly with increasing delay  $(T_p)$  of the microwave pulse relative to the start of the first rf pulse, and then  $I/I_0$  remains practically constant. When the microwave pulse is made to coincide with the center of the second rf pulse, the echo intensity increases to almost its initial value; then it decreases again and is restored anew when the microwave pulse is turned on after the appearance of the echo signal. Such a behavior of the echo signal is in definite correlation with the orientation of the nuclear moments relative to the quantization axis (z). We recall that after a 90° rf pulse the



FIG. 2. Nuclear spin echo signal intensity following the action of 90 and 180° rf pulses against the delay time ( $T_p$ ) of the start of the microwave pulse relative to the start of the rf pulse. P/P<sub>thr</sub> = 10 dB (P<sub>thr</sub>threshold excitation power of parametric spin waves), H<sub>0</sub> = 1050 Oe. Top (a-e)-diagrams illustrating the formation of the nuclear spin-echo signal following the action of 90 and 180° rf pulses.

nuclear magnetization is in a plane perpendicular to the z axis. In the time interval between the first and second pulses, the nuclear magnetization spreads out into a fan made up of individual components. Then during the time of the second  $(180^{\circ})$  pulse the plane in which the fan of nuclear moments is situated rotates 180° around the direction of the rf field H<sub>rf</sub>. At the instant of time corresponding to the center of the second pulse, this plane occupies a vertical position and the nuclear moments have a predominant projection along (parallel and antiparallel to) the z axis. After the second pulse, the fan of nuclear moments is again on a horizontal plane and the individual components converge to form a single nuclear magnetization, which induces the echo signal<sup>[9]</sup>. As seen from the diagram showing the formation of the echo signal in Fig. 2, the influence of the parallel pumping is minimal when the nuclear moments are directed along the z axis, and is maximal when they are directed perpendicular to the z axis. We can therefore propose on this basis that the parametric spin waves influence principally the transverse component of the nuclear magnetization.

The intensity of the observed echo signal is  $I = cm_0 exp(-t/T_2)$ , where  $m_0$  is the nuclear magnetization at t = 0,  $T_2$  is the transverse relaxation time, and c is a certain coefficient. Therefore the change of the echo intensity can in the general case be connected with the change of  $T_2$  under the influence of the parametric spin waves. In our experiments, a decrease of the echo intensity by a factor of e takes place at a microwave pulse power 10-15 dB above the instability threshold. Recognizing that the influence of the parametric pumping lasts only 1  $\mu$  sec, we find that the time of the transverse nuclear relaxation due to the parametric spin waves  $T_{2(pw)} = 1 \ \mu \text{ sec.}$  We note that without parametric pumping  $\mathbf{T}_2 = 10^3 \,\mu \, \text{sec}$ , so that the parametric spin waves cause the transverse nuclear relaxation to be accelerated by  $10^3$  times.

At the same time, we did not observe any influence of the parametric spin waves on the time of longitudinal relaxation (T<sub>1</sub>) or on the intensity of the so-called stimulated echo. To this end we used three 90° rf pulses. When the microwave pulse was turned on between the second and third rf pulses, i.e., in the time interval when only longitudinal components of the nuclear moments take part in the formation of the stimulated-echo signal, there is no influence of the microwave pumping. The corresponding figure is given in<sup>[8]</sup>. (We note that in the present article we deal throughout with the quantity I/I<sub>0</sub>, whereas in<sup>[8]</sup> we used the quantity  $\Delta I/I_0$ =  $1 - I/I_0$ ).

Figures 3 and 4 show plots of  $I/I_0$  against the microwave pulse power and against the external magnetic field.

#### 3. THEORY OF TRANSVERSE NUCLEAR RELAXATION

Transverse nuclear relaxation is due to low-frequency oscillations of the longitudinal component of the local field at the nuclei. We shall assume that the local field at a nucleus is due to the isotropic hyperfine interaction, which is described by a Hamiltonian  $\mathcal{H}_1 = A_i \mathbf{S}_i \mathbf{I}_i$ , where  $A_i$  is a constant,  $\mathbf{S}_i$  and  $\mathbf{I}_i$  are respectively the



FIG. 3. Dependence of the nuclear spin-echo signal intensity on  $P/P_{thr}$ -the ratio of the microwave pulse power to the threshold excitation power of the parametric spin waves,  $H_0 = 800 \text{ Oe}$ ,  $T_D = 30 \, \mu \text{sec}$ .

FIG. 4. Dependence of the nuclear spin-echo signal intensity on the external magnetic field  $H_0$ . Dark points-P/P<sub>thr</sub> = 7 ± 0.5 dB, light points-P/P<sub>thr</sub> = 5 ± 0.5 dB. The magnitudes of the wave vectors k of the parametric spin waves change from  $k \approx 5 \times 10^5$  cm<sup>-1</sup> at  $H_0 = 0$  to  $k \rightarrow 0$  at  $H_0 = 1800$  Oe.

electron and nuclear spin operators, and i is the number of the atom. Then, by definition, [4]

$$\frac{1}{T_2} = \frac{1}{2T_1} + \frac{A^2}{\hbar^2} \int_{-\infty}^{+\infty} \langle \{\delta S_i^z(t) \, \delta S_i^z(0)\} \rangle \, dt. \tag{1}$$

The integrand here is the correlation function for the fluctuations of the  $S^Z$  projections of the electron spin. The contribution from the spin-lattice relaxation  $(1/2T_i)$  in formula (1) can be neglected. If the  $S^Z$  components of the electron spin are expressed in terms of the spin-wave creation and annihilation operators, then the transverse nuclear relaxation can be represented as a result of two-magnon scattering of spin waves by the nuclear moment. In this process a spin wave with wave vector  $\mathbf{k}_1$  is scattered by the nuclear moment and is transformed into another spin wave having the same energy but with a wave vector  $\mathbf{k}_2$ . It is shown in<sup>[3,10,11]</sup> that

$$\frac{1}{T_2} = \left(\frac{A}{\hbar}\right)^2 \frac{\pi}{N^2} \sum_{\mathbf{k}_1 \neq \mathbf{k}_2} n_{k_1} (n_{k_2} + 1) \,\delta(\omega_{k_1} - \omega_{k_2}).$$
(2)

Here N is a normalization constant (on the order of the number of magnetic atoms in the sample), and  $n_{k_1}$  and  $n_{k_2}$  are the numbers of spin waves with wave vectors  $k_1$  and  $k_2$ . Formula (2) was obtained for the case when the Hamiltonian of the system does not depend explicitly on the time. In our case the sample is acted upon by a sufficiently powerful microwave field, but we will nevertheless still use relation (2), bearing in mind the fact that the corresponding corrections are actually small.

As is well known, in the case of parallel pumping a photon with the pump frequency  $\omega_p$  breaks up into two magnons with  $\omega_{k_1} = \omega_{k_2} = \omega_p/2$  and  $k_1 + k_2 = 0$ , if the microwave-field power exceeds a certain threshold value. The problem of the angular distribution of the wave vectors of parametric spin waves has not yet been completely solved<sup>[12]</sup>. We consider therefore two limiting cases.

A. Completely isotropic distribution of the wave vectors. Taking into account the damping of the parametric spin waves, i.e., replacing the  $\delta$  function by

$$\frac{1}{\pi} \frac{2\Delta\omega_k}{(\omega_{k_1} - \omega_{k_2})^2 + (2\Delta\omega_k)^2} = \frac{1}{2\pi\Delta\omega_k}$$

and assuming that the damping  $\Delta \omega_k$  does not depend on k, we get

$$\frac{1}{T_{z(\mathbf{pw})}} = \left(\frac{A}{\hbar}\right)^{z} \left(\frac{n}{N}\right)^{\frac{z}{2}} \frac{1}{2\Delta\omega_{k}} = \frac{\omega_{0}^{z}}{S^{z}} \left(\frac{n}{N}\right)^{\frac{z}{2}} \frac{1}{2\Delta\omega_{k}}.$$
 (3)

Here  $\omega_0$  is the NMR frequency and n is the total number of excited parametric waves.

**B.** Completely anisotropic distribution with respect to the wave vectors. We bear in mind here that the only spin waves to be excited are those propagating along a certain direction 1, i.e.,  $\mathbf{k}_1 = \mathbf{k}_l$  and  $\mathbf{k}_2 = \mathbf{k}_{-l}$ . Then the condition  $\mathbf{k}_1 \neq \mathbf{k}_2$  in formula (2) modifies expression (3) to read:

$$\frac{1}{T_{2(\mathbf{pw})}} = \frac{\omega_0^2}{S^2} \left(\frac{n}{N}\right)^2 \frac{1}{4\Delta\omega_h}.$$
(4)

As seen from (3) and (4), these two limiting cases differ only by a factor 1/2. (The result (4) has been written out in<sup>[7]</sup> with the following notation:  $\Delta \omega_{\mathbf{k}} = \gamma \Delta H_{\mathbf{k}}$  and  $n_{\mathbf{k}} = n/2$ .) Using the following values of the quantities in (3):  $\omega_0 = 2\pi \cdot 63 \cdot 10^6$ ,  $S = \frac{5}{2}$ ,  $n/N \sim 10^{-2}$  and  $\Delta \omega_{\mathbf{k}}$  $= \gamma \Delta H_{\mathbf{k}} = 5.0 \cdot 10^{6[13]}$ , we have  $1/T_2 \approx 10^5 - 10^6$  sec<sup>-1</sup>, which is in satisfactory agreement with experiment.

It is interesting to note that such effective scattering of spin waves by nuclei is typical precisely of spin waves and is practically nonexistent for homogeneous precession.

In connection with the foregoing results, notice should be taken of one more important feature, which distinguishes the interaction of nuclear moments with parametric spin waves from their interaction with thermal magnons. All the paramagnetic spin waves have the same energy, and therefore  $1/T_2$  is determined by the square of their total number. The contribution to  $1/T_2$  from the thermal magnons is made not by the square of their total number, but by the sum of the squares of the numbers of the spin waves with equal energy. It is therefore clear that even at an equal number of both thermal and parametric spin waves, the effect due to the parametric ones will be much larger than that due to the thermal ones, as was indeed observed in the experiment. To conclude the discussion of formula (3), we note that if the spin waves have a certain distribution in a frequency interval  $\omega_1 - \omega_2$  which is larger than 2  $\Delta \omega_{\mathbf{k}}$ , then the calculation of  $1/T_2$  is more complicated, but for an estimate it is possible to use formula (3) as before, replacing  $2 \Delta \omega_k$  by the difference  $\omega_1 - \omega_2$ .

A contribution to  $1/T_2$  can be made also by a process not connected with the scattering of the spin waves by the nuclei, but due to the change of the average value of the electron magnetization  $M_z$  at the start and end of the microwave pulse. It turns out that relation (3) is again valid for an estimate of this contribution, but it must be taken into account only during the short time interval during which the growth and relaxation of the parametric waves take place. Since in our experiment the duration of the microwave pulse is larger than the time of the transient processes, the main contribution to  $1/T_2$  is obviously made by the scattering of the parametric spin waves by the nuclear moment.

### 4. LONGITUDINAL NUCLEAR RELAXATION

The parametric spin waves can take part in all the processes of the longitudinal nuclear relaxation in which

thermal magnons take part. Let us consider some of them.

A. Single-magnon processes. In this case the nucleus absorbs or emits one spin wave. Usually this mechanism is forbidden because of the energy conservation law, since the spin-wave energy is as a rule larger than the energy of the flipping of the nuclear spin. But if account is taken of the damping of the spin waves, then such transitions are possible in the system. In this case, performing calculations analogous to those  $in^{[11,14]}$ , we get

$$\frac{1}{T_{i}} = \left(\frac{A}{\hbar}\right)^{2} \frac{S}{N} \sum_{k} \frac{2n_{k}\Delta\omega_{k}}{(\omega_{k} - \omega_{0})^{2} + \Delta\omega_{k}^{2}} \approx \left(\frac{A}{\hbar}\right)^{2} \frac{S2n}{N} \frac{\Delta\omega_{k}}{(\frac{1}{2}\omega_{p} - \omega_{0})^{2}}$$
(5)

Using the numerical values of the parameters given above, we have  $1/T_1 \approx 10 \text{ sec}^{-1}$ . We found that without microwave pumping we have in our sample  $1/T_1$ =  $10^2 \text{ sec}^{-1}$ , and therefore longitudinal relaxation due to parametric spin waves cannot be observed at a temperature ~  $140^{\circ}$ K. But at T =  $4.2^{\circ}$ K in our sample the experimental value is  $1/T_1 = 1 \text{ sec}^{-1}$ , and in this case relaxation due to parametric waves can be effective.

**B.** Two-magnon processes. In this process, the reversal of the nuclear moment is accompanied by absorption and emission of a spin wave, the difference in the energies of the radiated and absorbed spin waves being equal to the energy of reversal of the nuclear moment. These processes are allowed if conservation of the total angular momentum in the system is not required. Such conditions are encountered, for example, if the quantization axes of the nuclear and electron systems do not coincide, or if account is taken of the dipole-dipole interaction between the electron spins<sup>[15,16]</sup>. We present the result for the case when the quantization axes of the nuclear and electron moments differ by an angle  $\theta$ . The calculation is analogous to that performed in<sup>[15,16]</sup>:

$$\frac{1}{T_1} = \frac{4\pi}{\hbar} \frac{A^2}{N^2} \sin^2 \theta \sum_{\mathbf{h}_1, \mathbf{h}_2} n_{\mathbf{h}_1} (n_{\mathbf{h}_2} + 1) \delta(\hbar \omega_{\mathbf{h}_1} - \hbar \omega_{\mathbf{h}_2} - AS) \qquad (6)$$
$$= \frac{\sin^2 \theta}{\pi} \left(\frac{A}{\hbar}\right)^2 \frac{n}{N} \left(\frac{kT}{\hbar \omega_c}\right) \left(\frac{1/2\omega_p - \omega_1}{\omega_c}\right)^{1/2} \frac{1}{\omega_p}.$$

Here  $\omega_e$  is the exchange frequency,  $\hbar \omega_1$  is the energy of the gap in the spin-wave spectrum. Assuming  $\omega_e = 6 \cdot 10^{12}$ ,  $\omega_p = 2\pi \cdot 10^{10}$ ,  $\omega_1 = 2\pi \cdot 10^9$ ,  $n/N = 10^{-2}$  and T =  $10^{\circ}$ K, we have  $1/T_1 \approx (A/\hbar)^2 \sin^2\theta \cdot 10^{-15}$  [sec<sup>-1</sup>]. For thermal magnons under the same conditions, using the results of [15,16], we have  $1/T_1 \approx (A/\hbar)^2 \sin^2\!\theta \cdot 10^{-16}$ [sec<sup>-1</sup>]. Thus, the parametric waves can ensure a more effective relaxation mechanism than thermal waves if the quantization axes of the nuclear and electron systems do not coincide. For an yttrium garnet this mechanism is apparently not significant, since the hyperfine interaction is practically isotropic here ( $\theta \rightarrow 0$ ). It is perfectly possible, however, that allowance for the electronic dipole-dipole interactions will also give an effective relaxation mechanism for  $Y_3Fe_5O_{12}$ . (In analogy with the mechanism calculated in<sup>[16]</sup>, the only difference being that one of the magnons is not thermal but a parametric spin wave.)

**C.** Three-magnon processes. These processes are allowed both by the energy and by the momentum conservation laws. For thermal magnons, the results of

the calculations are given  $in^{[16]}$ . Estimates for threemagnon processes with participation of parametric spin waves shows that they are in general less effective, with the exception of the region of very low temperatures (<1°K). Thus, the mechanisms of longitudinal nuclear relaxation with participation of parametric spin waves can be more effective as a result of thermal magnons for single- and two-magnon processes in a wide temperature interval, and are not very effective for three-magnon processes.

It should be noted here that longitudinal relaxation in the processes under consideration will not be equivalent to the spin-lattice nuclear relaxation. The singlemagnon process of longitudinal nuclear relaxation will bring the nuclear system into equilibrium not with the lattice but with the reservoir of parametric magnons. This corresponds to relaxation of the nuclear system to a state with a spin temperature much higher than the lattice temperature.

For two-magnon processes, the system of nuclei exchanges with the lattice not an energy quantum equal to the energy of the nuclear flip ( $\hbar \omega_0$ ), but a quantum  $\hbar \omega_k \pm \hbar \omega_1$ , and consequently the ratio of the populations of the nuclear levels will not equal the Boltzmann factor  $\exp(-\hbar \omega_0/kT)$ . Accordingly, the temperature of the nuclear system will differ from the lattice temperature. This question is discussed in greater detail by Oguchi and Keffer<sup>[17]</sup>.

## 5. SINGULARITIES OF OBSERVING NUCLEAR ECHO IN THE PRESENCE OF MICROWAVE PUMPING

So far we have considered the influence of parametric spin waves on the nuclear relaxation. Let us stop now to discuss the influence of microwave pumping on the frequency and intensity of the echo signal. Essentially in this experiment we deal with a new type of electronnuclear double resonance in magnetically ordered crystals. Double resonance due to interaction of nuclear moments with homogeneous precession has been reported earlier<sup>[18,19]</sup>. In our case we are dealing with interaction between the nuclear moments and the spin waves.

Excitation of parametric spin waves leads to a decrease of the mean value of the longitudinal component of the electron magnetization and to its modulation at the pump frequency  $\omega_p$ . This results in a decrease of the longitudinal component of the local field at the nucleus and hf modulation of this component. The influence of modulation can be neglected, since these oscillations average out effectively during the lifetime of the nucleus at the level, in analogy with the averaging of the local field in the case of rapid molecular motion or in experiments with rapid rotation of the sample in an inhomogeneous magnetic field. But the change of the average value of the local field gives a shift of the resonant frequency. Excitation of spin waves whose number is  $n = N \times 10^{-2}$  leads to a change of  $M_Z$  by an amount  $\Delta M_z \approx M_z \times 10^{-2}$ , and to a change of the resonant frequency  $\Delta f_0 = f_0 / 10^{-2}$  by approximately 0.6 MHz. This shift towards lower frequencies greatly exceeds the NMR line width  $\sim 0.3$  MHz (Fig. 5).

However, this shift occurs only during the time of application of the microwave pulse, i.e., during  $\sim 1 \ \mu$  sec. In experiments on nuclear echo, the line



FIG. 5. Dependence of the nuclear spin-echo signal intensity on the carrier frequency of the rf pulses. The sample is acted upon by a 90° (pulse duration  $\tau_1 = 4 \,\mu\text{sec}$ ) and a 180° ( $\tau_2 = 8 \,\mu\text{sec}$ ) rf pulse. Solid line-without application of microwave power, dash-dot-P/P<sub>thr</sub> = 7 dB, dashed-P/P<sub>thr</sub> = 12 dB. The microwave pulse is applied during the time of action of the second rf pulse and its T<sub>p</sub> was chosen to ensure maximum echo signal (see Fig. 2); H<sub>0</sub> = 1050 Oe.

FIG. 6. Dependence of the echo signal intensity following the action of rf pulses of duration 4 and 10  $\mu$ sec (black points) and rf pulses of duration 4 and 6  $\mu$ sec on the position of the microwave pulse.  $\Delta T_p$ -time between the start of the second rf pulse and the start of the microwave pulse, P/P<sub>thr</sub> = 7.5 dB, H<sub>0</sub> = 1050 Oe.

shift can appear if the rf pulse coincides with the microwave pulse. In our experiments, this shift is not observed, but a broadening and distortion of the shape of the NMR line are observed when the microwave pulse is applied at the center of the 180° rf pulse. It is necessary to bear in mind here that besides the line shift there occurs also a dynamic broadening by an amount  $1/\pi T_{2(pw)} \approx 0.3$  MHz, and the observed dependence of the signal intensity on the frequency is a superposition of several effects.

An analysis of the line shifts and of the line shape is very complicated in the pulsed version of the experiment with which we are dealing. We call attention to the increase in both the integral and peak intensities of the NMR signal with increasing microwave power. We are inclined to regard this effect as only illusory and resulting from the fact that we are observing a certain superposition of a shifted NMR with an unshifted one. Incidentally, other explanations are also possible here.

The behavior of the echo-signal intensity (Figs. 3 and 4) when the microwave pulse does not coincide with the center of the 180° rf pulse, can be readily explained qualitatively on the basis of the relation I =  $cm_0exp(-t/T_{2(pw)})$ . With increasing microwave power, the number of excited spin waves n increases, as a result of which  $T_{2(pw)}$  decreases and consequently the signal intensity drops. Similarly, with increasing H<sub>0</sub> a decrease takes place in  $\Delta \omega_k^{[13]}$ , as a result of which  $T_{2(pw)}$  decreases and a decrease of the signal is observed. We note that these data can in principle be used to determine the dependence of  $\Delta \omega_k$  on H<sub>0</sub> or on the value of the wave vector, since spin waves with different **k** are excited with changing field H<sub>0</sub>. More complicated processes occur when the microwave pulse coincides with the rf pulse, especially with the second rf pulse. Here the echo-signal intensity depends not only on the rate of transverse nuclear relaxation but also on the angle of rotation of the plane of the fan of the nuclear magnetization relative to the z axis. Since the angle of rotation depends on the duration and on the power of the microwave pulses, the intensity of the echo in the presence of the microwave pumping should depend in some manner on the duration of the rf pulses.

In Fig. 6 we show more detailed data for the echosignal intensity, when the microwave pulse "passes" through the second rf pulse. The black circles show the results of measurements for a second-RF-pulse duration  $\tau_2 = 10 \ \mu \text{sec}$  (close to the 180° pulse), and the white circles are for  $\tau_2 = 6 \ \mu \text{sec}$  (close to the 90° pulse).

Qualitatively, the difference in the behavior of the signal can be explained in the following manner. When the microwave pulse passes through a 180° rf pulse, the plane of the fan is directed vertical in the center of the 180° pulse and the influence of the fast transverse relaxation is minimal. Therefore the signal is restored to its initial value  $I_0$  (without microwave pumping). We call attention to the fact that a fine structure, similar to an oscillation, is observed on the upper part of the plot, and the intensity of the signal at the center is even slightly higher than the initial intensity ( $I/I_0 > 1$ ). These facts have not yet been unambiguously explained.

For 90° rf pulses, the conditions for the formation of the echo signal differ from the scheme given in Fig. 2 of the present article<sup>[20]</sup>. In this case the influence of transverse relaxation is significant during the time of the entire duration of the second pulse. Accordingly, we see in Fig. 6 that the intensity of the echo signal is not restored to its initial level. Without stopping to analyze these effects in greater detail, we wish to emphasize that the experiments described here can be useful for a detailed study of the dynamics of motion of nuclear magnetization under the influence of radiofrequency pulses. It is possible that similar experiments would make it possible to trace the motion of the vector of nuclear magnetization in different methods of echosignal excitation.

In conclusion, we note that the described effects were investigated in a ferrimagnetic compound, but similar investigations are possible also in antiferromagnets in those cases where parametric spin waves can be excited.

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