Variation of the spin-wave spectrum with interaction of magnons

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In a specimen of the antiferromagnetic crystal $OsMnF_3$, the variation of the characteristic frequency of magnons with a definite value $k = k_1$ of the wave number has been observed on excitation of magnons with $k = k_2$. The magnons were excited parametrically, by microwave pumping. The density of the parametrically excited magnons was of order 10^{17} cm^{-3}. The relative change of the characteristic frequency (shift of the magnon spectrum) is $\sim 10^{-5}$. The variation of the characteristic frequency is recorded and measured by observation of transition processes in the system of parametrically excited magnons. The frequencies of the magnons under consideration were 10.5 and 17.5 GHz ($k_1 \sim k_2 \sim 10^5$ cm^{-1}); the specimen temperature was $T = 1.6$ K. The amplitude of the four-magnon interaction that causes the spectral shift was determined: $T_{12}/2\pi \approx -10^{-12}$ Hz cm^3.

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INTRODUCTION

In 1930, F. Bloch developed a theoretical representation of spin waves as an ideal gas of noninteracting elementary excitations.[1] In the later development of the theory, interactions of spin waves have been intensively studied. Interaction of spin waves with each other and with lattice oscillations has two basic consequences:

1) They determine the finite lifetime of a magnon and the length of the free path[2];

2) interaction of the attraction or repulsion type leads to a change of the spin-wave spectrum in a strongly excited crystal.[3,4]

Probability estimates have been measured experimentally or derived only for magnon interaction processes that give results of the first type. In experiments on measurement of the relaxation frequency of spin waves,[5,6] data are obtained on the sum of all such interactions. In experiments on scattering of magnons of one frequency by magnons of another frequency,[7,8] what is determined is the probability of a relaxational interaction (i.e. on with a result of the first type) of two magnons with definite values of the quasimomenta. For antiferromagnetic $CsMnF_3$, it has been ascertained[8] that the basic process of interaction of two magnons of the quasiferromagnetic branch of the spectrum, with frequencies of about 20 GHz (that is, "average" magnons of a heat reservoir at $T \approx 1-2$ K), is a process of fusion of two magnons into a phonon.

The change of the spectrum caused by interaction of spin waves has not yet been observed experimentally, although this phenomenon has been elucidated in a number of theoretical papers (see Refs. 3 and 4 and others). In the present paper, we describe an observation of the effect of change of the spectrum of the magnons by nondissipative interaction of them with each other; we describe the method and give experimental data on the spectral shift and on the amplitude of the interaction that causes it.

As follows from the theoretical investigations,[4] among all the forms of three- and four-magnon interactions, the only processes that lead to a change of the spectrum are those in which the two initial magnons either do not change their quasimomenta or exchange them. The contribution to the change of the dispersion law from processes that involve participation of a larger number of magnons is small in comparison with that considered.

The Hamiltonian of the interaction described is determined by the expression

$$\mathscr{H}_{int} = \sum_{\mathbf{k},\mathbf{k}'} T_{\mathbf{k},\mathbf{k}'} c_{\mathbf{k}} c_{\mathbf{k}}^{+} c_{\mathbf{k}'} c_{\mathbf{k}'}^{+}, \tag{1}$$

where $c_{\mathbf{k}}$ and $c_{\mathbf{k}}^{+}$ are the creation and annihilation operators for magnons with wave vector $\mathbf{k}$, and where $T_{\mathbf{k},\mathbf{k}'}$ is the amplitude of the interaction. It must be noted that this process makes no contribution to the relaxation of magnons, since it does not change the number of magnons with a definite value of the quasimomentum; that is, it is dissipationless. The spin-wave spectra in the unexcited crystal and in a crystal with a large number of magnons ($\omega_{\mathbf{k}}$ and $\tilde{\omega}_{\mathbf{k}}$) are connected by the relation[4]

$$\tilde{\omega}_{\mathbf{k}} = \omega_{\mathbf{k}} + 2 \sum_{\mathbf{k}'} T_{\mathbf{k}\mathbf{k}'} n_{\mathbf{k}'}; \tag{2}$$

here $n_{\mathbf{k}'}$ is the number of magnons with quasimomentum $\mathbf{k}'$.

In the present paper, the crystal investigated was of antiferromagnetic $CsMnF_3$, with anisotropy of the "easy plane" type. The spin-wave spectrum of the low-frequency branch in an unexcited crystal of this substance has the form

$$\omega_{\mathbf{k}} = g[H_\Delta^2/T_n + H^2 + \alpha^2 k^2]^{1/2}, \tag{3}$$

where g is the gyromagnetic ratio, $H_\Delta^2 = 6.3$ kOe^2 is a constant that describes the interaction of the electronic and nuclear spins, T_n is the temperature of the nuclear-spin subsystem, H is the external magnetic field, and

α is the exchange constant.

The quantity

$$\Delta=2\sum_{\mathbf{k}'}T_{\mathbf{k}\mathbf{k}'}n_{\mathbf{k}'}$$

represents the nonlinear frequency shift (NFS) of magnons with quasimomentum **k**.

The amplitude $T_{\mathbf{k}\mathbf{k}'}$, for antiferromagnets with anisotropy of the easy-plane type, has been calculated theoretically by L'vov and Shirokov[9]:

$$T_{\mathbf{k}\mathbf{k}'}=-\frac{g^2\hbar}{8}\frac{H_E}{M_0}\frac{g^2(H_\Delta^2/T_n+4H^2)}{\omega_{\mathbf{k}}\omega_{\mathbf{k}'}} \tag{4}$$

(H_E is the exchange field; M_0 is the sublattice magnetization).

METHOD

The basis of the proposed method of studying NFS is the phenomenon of parametric excitation os spin waves (see, for example, Refs. 4 and 5). In our setup, the crystal is subject to the action of two microwave pumps with frequencies ω_{p1} and ω_{p2}, which parametrically excite in the specimen magnons with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ and frequencies

$$\text{a) } \tilde{\omega}_{\mathbf{k}_1}=\omega_{p1}/2, \quad \text{b) } \tilde{\omega}_{\mathbf{k}_2}=\omega_{p2}/2. \tag{5}$$

The number of these magnons greatly (by a factor of about 10^6) exceeds the thermal background of magnons with such wave vectors. We shall designate magnons parametrically excited by the pump with frequency ω_{p1} by the abbreviation PM1, and magnons that originate in the action on the crystal of the second pump by PM2.

In accordance with the discussion in the Introduction, the presence of magnons PM2 changes the spectrum of magnons with wave vector $\mathbf{k}_1$ by the amount $\Delta_2=2T_{12}N_2$, where N_2 is the number of PM2. Therefore if a stationary state of absorption of the microwave power of the first pump has been established in the crystal, i.e. a stationary level of the number of magnons PM1 (N_1), and if, as a result of switching the second pump on or off, there occurs a rapid change of the value of N_2 from zero to the stationary level (or the opposite), then the magnons PM1 no longer satisfy the condition for parametric resonance [equation (5) a)], and a transitional process should be observed, in the course of which nonresonance magnons decay, while the number of magnons with the resonance value of the quasimomentum increases.

A schematic diagram of the experiment set up according to this principle is given in Fig. 1. In order to produce at the specimen two high-frequency magnetic fields with frequencies $\omega_{p1}/2\pi=21.36$ GHz and $\omega_{p2}/2\pi=35.1$ GHz, the crystal was placed in a bimodal resonator in

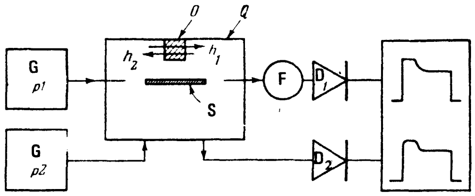

FIG. 1. Schematic diagram of the experiment. G_{p1}, first pump generator (21.36 GHz); G_{p2}, second pump generator (35.1 GHz); D_1 and D_2, microwave-signal detectors; F, filter; S, copper strip of strip resonator; h_1 and h_2, microwave magnetic pumping fields; O, specimen; Q, resonator.

the form of a rectangular metallic cavity, parallel to one of whose walls was placed a strip of copper foil. The cavity dimensions were so chosen that the spatial oscillation of type H_{012} has frequency ω_{p2}. The copper strip and the wall of the cavity form a strip resonator.[10] The size of the strip determines the frequency ω_{p1} of the strip resonator. The channel for recording of the first pump contains a filter that delays the signal of frequency ω_{p2}. The waveguide of the signal of the second pump is beyond the limits for the signal of frequency ω_{p1}. This makes it possible to observe the signals of each of the pumps separately by means of the crystal detectors D_1 and D_2. The crystal detectors produce a voltage proportional to the power of the signals incident upon them. The specimen was at temperature 1.6 K and was located at a loop of the distribution of both microwave fields. The condition for parametric excitation of spin waves in the antiferromagnet, $\mathbf{H}\parallel\mathbf{h}\perp C^6$, was satisfied. Here C^6 is a six fold symmetry axis of the crystal under investigation, and **h** is the microwave magnetic field.

It is possible to excite parametrically in the specimen pairs of magnons with a frequency equal to half the pumping frequency and with oppositely directed wave vectors. The wave vector of the parametrically excited magnons (PM) is determined by the condition $\tilde{\omega}_{\mathbf{k}}=\omega_p/2$ and, for a given value of ω_p, varies, depending on the magnetic field H, over the interval 0 to 10^5 cm^{-1}. Excitation of PM occurs when the pumping field h exceeds the threshold value h_c; it is registered by the microwave power absorption in the specimen. In the antiferromagnets $CsMnF_3$ and $MnCO_3$, parametric excitation is of the "hard" kind[5]: an intense increase of the number of PM and of the absorbed power begins a certain time after the switching on of the microwave pump, and then these quantities rapidly reach stationary values; as a result, an oscillogram of the pulse of power transmitted through the microwave resonator containing the specimen has the form of a rectangular pulse with a step.

The spin system possesses a quasicontinuous spectrum of elementary excitations; therefore the nonlinear shift of the magnon frequency does not lead to a limiting of the amplitude of the parametrically excited spin waves, since there are always magnons that are in exact parametric resonance with the pump.

In a number of papers[4,11] it has been shown that the limiting of the number of parametrically excited magnons and the establishment of a stationary state occur in consequence of a phase shift of the pair of PM with respect to the pump. By the phase of a pair of parametrically excited magnons, with wave vectors **k** and **-k** and complex amplitudes $C_{\mathbf{k}} = |C_{\mathbf{k}}| \exp(i\varphi_{\mathbf{k}})$ and $C_{-\mathbf{k}} = |C_{-\mathbf{k}}| \exp(i\varphi_{-\mathbf{k}})$, is understood the quantity $\psi_{\mathbf{k}} = \varphi_{\mathbf{k}} + \varphi_{-\mathbf{k}}$, which determines the power absorbed from the pump:

$$W = 2n_k hV \sin\psi_k; \qquad (6)$$

here V is the coefficient of coupling of the PM with the pump.

Figure 2 shows oscillograms of the power transmitted through the resonator from the pumps (the upper beam is the signal of the first pump, magnons PM1; the lower, of the second pump, magnons PM2); the moments of rapid change of N_2 are marked. The increase of N_2 occurs a certain time after switching on of the second pump, at the moment when a step appears on the oscillogram drawn by the lower beam. This process is marked with the label t_1. The decrease of N_2 follows after switching off of the second pump, in the process of decay of PM2 to the level of the thermal background. This moment is marked with the label t_2. In the vicinity of the moments of change of N_2 can be seen disturbances of the stationary state of absorption of power by magnons PM1 (the upper beam).

In order to understand and analyze the observed transitional processes, it is necessary to turn to the equations that describe the evolution of the number and phase of the PM (equations 3.6 in Ref. 4). In the simplest case of excitation by a single pump of magnons of only a single wave number k_1, these equations have the form

$$\tfrac{1}{2}\dot{N}_1 = N_1(-\gamma_1 + hV\sin\psi_1), \qquad (7a)$$

$$\tfrac{1}{2}\dot{\psi}_1 = \tilde{\omega}_{k_1} - \tfrac{1}{2}\omega_{p1} + hV\cos\psi_1 + S_{11}N_1, \qquad (7b)$$

where N_1 and ψ_1 are the number and phase of magnons with wave number k_1; h is the pumping field, V is the coefficient of coupling of the magnons with the pump; S_{11} is a coefficient describing the interaction of pairs of magnons PM1, which leads to a phase shift of the PM with respect to the pump; $2\gamma_1$ is the angular frequency of relaxation of the spin waves. Equation (7a) describes the passage of energy into and out of the magnon system; (7b) describes the establishment of the phase of the magnons as it depends on the detuning of their characteristic frequency with respect to the half-frequency of the pump.

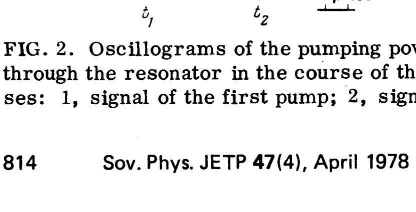

FIG. 2. Oscillograms of the pumping power transmitted through the resonator in the course of the transitional processes: 1, signal of the first pump; 2, signal of the second pump.

The stationary values of N_1 and ψ_1 are determined by the conditions $\dot{N}_1 = \dot{\psi}_1 = 0$ and the resonance condition $\tilde{\omega}_{k1} = \omega_{p1}/2$. With parametric excitation of PM1 and PM2 we get according to (2)

$$\tilde{\omega}_{k_1} = \omega_{k_1} + 2T_{11}N_1 + 2T_{12}N_2. \qquad (8)$$

In the vicinity of the moments marked t_1 and t_2 on the oscillogram, there occurs a change of $\tilde{\omega}_{k1}$ by the amount $\Delta_2 = 2T_{12}N_2$. This leads to a violation of the stationary condition $\dot{\psi}_1 = 0$, the phase ψ_1 deviates from the stationary value, and a change of the absorbed power W occurs in accordance with (6). The stationary value of ψ_1 is enclosed between 0 and $\pi/2$; therefore an increase of W observed in the vicinity of the moment t_2 is possible when $\psi_1 > 0$. Because here the number of magnons PM2 decreases, therefore $\Delta_2 < 0$; that is, the coefficient T_{12} is negative.

A similar transitional process in a PM system was observed by us earlier[11] after a rapid change of the pumping frequency by an amount $\delta\omega_p$. Then the stationary condition $\dot{\psi} = 0$ is violated by the amount $-\delta\omega_p/2$, and a transitional process is also observed (see Fig. 10 in Ref. 11). Therefore in order to measure the NFS Δ_2, we subjected the magnons PM1 to a simulation action where in the pumping frequency was detuned from its original value by the amount $\delta\omega_p$. This change occurred over a time interval approximately equal to the lifetime of PM2, $\tau_{M2} = 1/2\gamma_2 \sim 1$ μsec.

The change of pumping frequency was accomplished by a pulsed voltage with regulated duration of the front, fed to the repeller plate of the klystron microwave generator. The time τ_{M2} was determined from the duration of the transitional process—beats in the PM2 system (see Ref. 11)—that occurred after rapid switching of the pumping frequency by the amount $\delta\omega_p \gg hV$.

If the change of pumping frequency $\delta\omega_p = -2\Delta_2$, then the initial conditions, and also the change in time of the external parameters for equations (7), will be the same as for switching on of the second pump at the moment t_2. Consequently, when $\delta\omega_p = -2\Delta_2$ the oscillograms of the power transmitted through the resonator in the vicinity of the moment t_2 and of the moment of change of the pumping frequency should be the same. By observing both of the transitional processes and choosing the change of pumping frequency $\delta\omega_p$ such that the oscillograms of the processes were the same, we determined Δ_2.

We remark that the value of $\delta\omega_p$ chosen in the above-described manner, within the limits of experimental error, does not change when the duration of the front of the pulse that shifts the generator frequency is changed by two or three times τ_{M2}.

The observed values of NFS are of order $2\pi\cdot 0.1$ MHz; that is, about $10^{-5}\omega_{k1}$. It should be noted that a slight heating of the nuclear subsystem (by about 10^{-3} K) can cause the same shift of the spectrum according to (3). Furthermore, a decrease of the sublattice magnetization as a result of excitation of spin waves will cause a change of the energy of interaction of the electronic and nuclear subsystems and will lead to a shift of the spectrum. In order to estimate these effects, it is necessary to know the spin-lattice relaxation times of the nuclear and electronic subsystems, τ_{nl} and τ_{el}. These times are known to within an order of magnitude and only for uniform precession of the nuclear magnetization[12] and for electronic spin waves with $k \approx 10^5\ \mathrm{cm}^{-1}$.[8] Estimation by use of these data shows that the shift of the spectrum caused by change of electron-nucleus interaction is smaller by one to two orders of magnitude than the observed.

Since these estimates are very approximate, we conducted the following control experiment. By means of a small coil wound around the specimen, the specimen was subjected to the influence of a radiofrequency field at the NMR frequency of the Mn^{55} nucleus. This produced a heating of the nuclear system to a temperature ~20 K, which was monitored by the shift of the AFMR frequency (ω_k with $k=0$) according to formula (3). Under these conditions, because of the decrease of the polarization of the nuclei, the contribution of interaction of electronic and nuclear spins to the frequency of the magnons of the electronic spin system decreases by a factor of more than ten. The experiment showed that the heating of the nuclear system did not lead to a significant change of the oscillogram of the observed transitional process. Consequently the heating of the nuclear-spin subsystem that occurs during excitation of PM, and the decrease of electron-nucleus interaction, do not interfere with the observation and measurement of the NFS of the magnons.

The remaining constants that determine the spectrum (g and α) do not depend on temperature. Increase of the field H in the specimen because of decrease of the demagnetizing field of the specimen on excitation of PM would shift the spectrum in the direction of an increase of $\tilde{\omega}_{k1}$. Therefore the prinicpal influence on the course of the observed transitional process is exerted by the NFS.

RESULTS

The results of measurements of the NFS Δ_2 for several values of the magnetic field are shown in Fig. 3. The abscissa of this graph is the value of $(P/P_c-1)^{1/2}$, where P is the power of the second pump and P_c is the corresponding threshold power; this quantity is proportional to the number of magnons PM2[13,14]:

$$N_2=A(H)\frac{2\gamma_2}{H^2}\left(\frac{P}{P_c}-1\right)^{1/2}. \qquad (9)$$

Here $2\gamma_2$ is the angular frequency of relaxation of PM2, and $A(H)$ is a coefficient describing the dependence of χ'' on the magnetic field.

As is seen from Fig. 3, the value of the NFS Δ_2 is proportional to the number N_2 of magnons producing it.

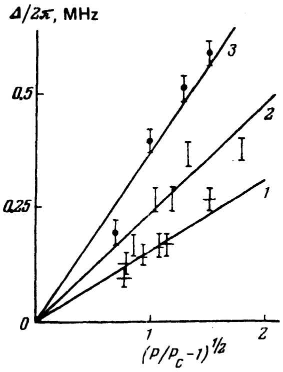

FIG. 3. Results of representative measurements of nonlinear frequency shift (the absolute value of the effect is shown): 1, $H=1.68$ kOe; 2, $H=2.24$ kOe; 3, $H=2.8$ kOe.

By measuring the value of the power absorbed in the crystal,

$$W=N_2\hbar\tilde{\omega}_{k2}2\gamma_2 \qquad (10)$$

and using the known value of $2\gamma_2$, one can determine N_2 and consequently T_{12}. For a specimen of volume 1 cm^3 in a field of 1.10 kOe, we get: $T_{12}/2\pi=-10^{-12}$ Hz $\pm$ 50%. This agrees approximately with the value obtained theoretically [formula (4)]: $T_{12}/2\pi=-3\cdot10^{-12}$ Hz.

In order to determine the variation of the coefficient T_{12} with magnetic field, one must convert the slope of the straight lines of Fig. 3 in accordance with formula (9). We get

$$T_{12}=\Delta_2/2N_2\propto\Delta_2H^2/A(H)\gamma_2.$$

The function $T_{12}(H)$ obtained by use of the experimental functions $A(H)$ and $\gamma_2(H)$ is shown in Fig. 4. There also are shown the results of measurement of the effect of magnons PM1 on the frequency of magnons PM2; that is, the values of T_{21}. Here the ratio of the lifetimes of the magnons is less favorable for clear manifestation of the effects, and in consequence the accuracy of the measurements is somewhat worse than in the first case; $\tilde{\omega}_k$ does not undergo its whole change during the time of decay of the "measuring" magnons. The coincidence of the absolute values of T_{12} and T_{21}, as well as their ab-

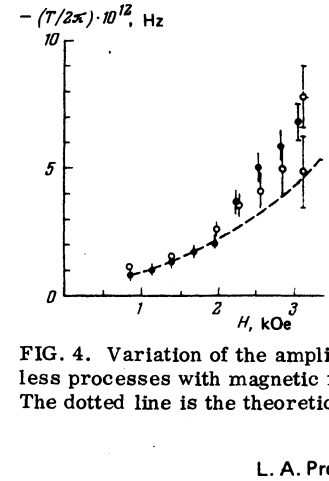

FIG. 4. Variation of the amplitude of four-magnon dissipationless processes with magnetic field: ●, values of T_{12}; ○, of T_{21}. The dotted line is the theoretical relation according to Ref. 9.

solute values, was established with accuracy 50%, since use is made here of measurements of the power of the generators, of the coefficients of coupling of a resonator with waveguides, and of the absolute values of τ_{M1} and τ_{M2}, which can not be done with high accuracy.

The dotted line in Fig. 4 shows the theoretical function

$$T_{12}(H)=T_{21}(H)\propto H_\Delta^2/T_n+4H^2$$

according to formula (4). Qualitative agreement of the theoretical and experimental functions is observed.

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NMR in hematite crystals with tin impurity

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NMR of Fe^{57} nuclei in the domain walls of hematite crystals with 0.8 wt.% tin impurity is investigated by the autodyne method. On the whole, the crystals remain weakly ferromagnetic down to 4.2 K, but an analysis of the temperature dependence of the NMR line shape and of the specific weakly ferromagnetic moment indicates that the most perfect sections of well-annealed crystals become antiferromagnetic at $T<200$ K. Quenching, which increases the defect content, makes practically the entire volume of the crystals ferromagnetic when they are cooled. The results point to an exceedingly strong sensitivity of the Morin temperature to the distributions of the defects and of the internal stresses.

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It follows from earlier studies[1-4] that NMR of Fe^{57} nuclei in hematite is observed only in the weakly ferromangetic phase, where conditions exist for high values of the gain, and the signal is due only exclusively to the nuclei in the domain walls.

Analyzing the line shape obtained by the autodyne method, Hirai *et al.*[4] have concluded that hematite crystals have walls of two types which lead to the appearance of NMR signals with opposite phases. The first type of wall is easily mobile, exists in well-annealed crystals, and is characterized by a small rigidity constant α in the equation of motion of the domain wall:

$$\mu\ddot{x}+\beta\dot{x}+\alpha x=2MH,$$

where μ is the effective mass of the wall, β is the damping constant, M is the weakly ferromagnetic moment, and H is the magnetic field.

For mobile walls walls, the domain-wall resonance frequency ω_W lies below the NMR frequency ω_N;

$$\omega_N>\omega_W=(\alpha/\mu)^{1/2}.$$

Mobile walls are easily annihilated in constant magnetic fields of the order of 10 Oe.

The second type of wall is characterized by high values of α and correspondingly by high domain-wall resonance frequency, so that the condition $\omega_W>\omega_N$ is satisfied for it. Such walls vanish in stronger magnetic fields.

The difference between the phases of the signals from pinned and free walls is due to the fact that at resonance the absorbed energy is proportional to

$$1+m\chi_n'$$

(χ_n' is the real part of the nuclear susceptibility), where the parameter m depends on the ratio of the frequencies

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