PARAMETRIC EXCITATION OF SPIN WAVES IN FERROMAGNETIC $MnCO_3$

V. V. KVEDER, B. Ya. KOTYUZHANSKIĬ, and L. A. PROZOROVA

Institute of Physics Problems, USSR Academy of Sciences

Submitted June 22, 1972

Zh. Eksp. Teor. Fiz. 63, 2205–2213 (December, 1972)

Parametric excitation of spin waves in the low-frequency branch of the spectrum is studied in antiferromagnetic $MnCO_3$ on parallel orientation of a high-frequency and static magnetic fields. The experiments are carried out at a frequency $\nu_p = 36$ GHz with prolonged (≈ 15 msec) pulses and at liquid helium temperatures. The appearance and disappearance of absorption in the sample is of discontinuous "hard" character. The results of the experiments are consistent with the assumption that a certain relaxation mechanism exists ($\Delta\nu_1$) which switches on at a certain magnon density n_{k1}. The dependences of $\Delta\nu_1^0$ and of stationary relaxation $\Delta\nu_2$ on temperature and magnetic field strength are studied in detail.

INTRODUCTION

PARAMETRIC excitation of spin waves in antiferromagnets by parallel pumping was observed in $CsMnF_3$ and $MnCO_3$, which have an anisotropy of the "easy-plane" type[1-4], and also on the "easy-axis" $CuCl_2 \cdot 2H_2O$ in fields H stronger than the flipping field[5].

A theoretical analysis of this process[6] led to the following results: There exists a certain threshold value of the microwave field h_c, above which the density n_k of spin waves with wave vector k begins to increase, first exponentially, and then assumes a stationary value. The wave vector of the excited spin waves depends on the applied field H and is determined from the dispersion law for the low-frequency branch of the spectrum:

$$(\nu_k/\gamma)^2 = H(H+H_D) + H_\Delta^2 + \alpha_z^2 k_z^2 + \alpha_\perp^2 k_\perp^2,$$

where $\nu_k = \nu_p/2$ is the frequency of the excited spin waves, ν_p is the pump frequency, H_D is the Dzyaloshinskii field, γ is the gyromagnetic ratio, H_Δ is the gap determined by the hyperfine interaction, and α_z and $\alpha_\perp$ are exchange constants.

In the case when $\nu_p \ll \nu_{20}$, where ν_{20} is the frequency of the high-frequency branch of the antiferromagnetic-resonance (AFMR) spectrum, the expression for the threshold field is

$$h_c = \nu_p \Delta\nu_k / \gamma^2 (2H + H_D), \quad (1)$$

and the magnon density in the exponential-growth section can be represented in the form

$$n_k = n_{k0} \exp\{\Delta\nu_k (h/h_c - 1)t\}, \quad (2)$$

where $\Delta\nu_k$ is the relaxation frequency of the spin waves of the low-frequency branch of the spectrum with wave vector k (to simplify the notation, the subscript k of $\Delta\nu_k$ will henceforth be omitted), n_{k0} is the thermal equilibrium density of the spin waves, h is the microwave field acting on the sample, and t is the time of action of the microwave power.

It follows from (2) that if we neglect the pump instability the spin-wave growth can be made arbitrarily slow by decreasing the excess of power above threshold. Nonetheless, experiment has shown (Fig. 2b of[4]) that at any small excess above threshold the spin-wave excitation has a jumplike "hard" character.

The purpose of the present study was to investigate thoroughly the relaxation and kinetics of the onset of spin waves following parametric excitation.

PROCEDURE

We used in the experiment the setup described in detail in[4]. The only difference was that the klystron generator was not swept, but operated in the long-pulse regime.

A single-crystal $MnCO_3$ sample measuring $2 \times 2 \times 0.3$ mm was secured with BF-6 glue to the bottom of a cylindrical resonator ($Q \approx 10\,000$) in the antinode of the magnetic field of the H_{012} mode. The static magnetic field was produced with a laboratory electromagnet. The two fields H and h were parallel and were in the principal plane of the crystal. The resonator was filled with superfluid helium to keep the sample from overheating. The microwave pulse passing through the resonator was detected with a crystal and fed to an oscilloscope. The pump frequency ν_p was ~ 36 GHz. The spin-wave excitation was revealed by the appearance of a distortion in the form of a break. Figure 1 show oscillograms of the initial pulse and of a pulse distorted by beyond-threshold absorption.

In a number of experiments we applied to the resonator microwave pulses with a waveform such that the power decreased smoothly with time (Fig. 2). This made it possible to study the transient process not only at the onset of the parametrically excited spin waves, but also at their vanishing. The variation of the pump frequency during the time of such a pulse can be neglected, since it does not exceed 2 MHz, and it follows from simple calculation that the relative change during the magnon lifetime ~ 1 μsec is < 0.2 kHz, i.e., $\ll \Delta\nu$.

The instantaneous value of the microwave power in the resonator was measured by the procedure described in[4]. The absolute measurement accuracy was $\approx 20\%$, whereas the relative change of the power was registered accurate to 5%. The experiments were performed in

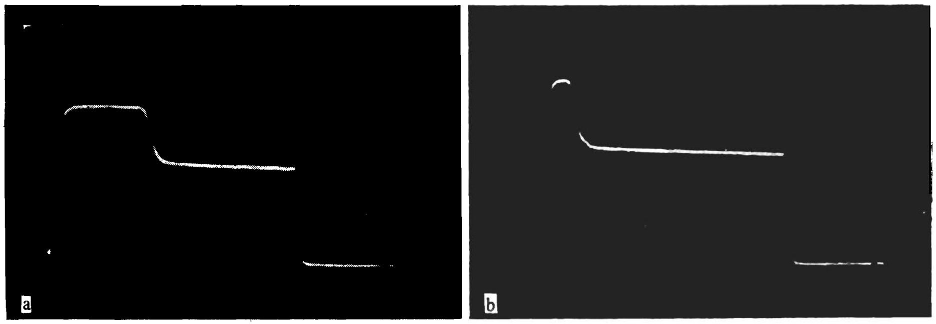

FIG. 1. Oscillograms of rectangular pulse passing through the resonator in the presence of beyond-threshold absorption in the sample: a—τ_1 = 80 μsec; b—τ_1 = 20 μsec.

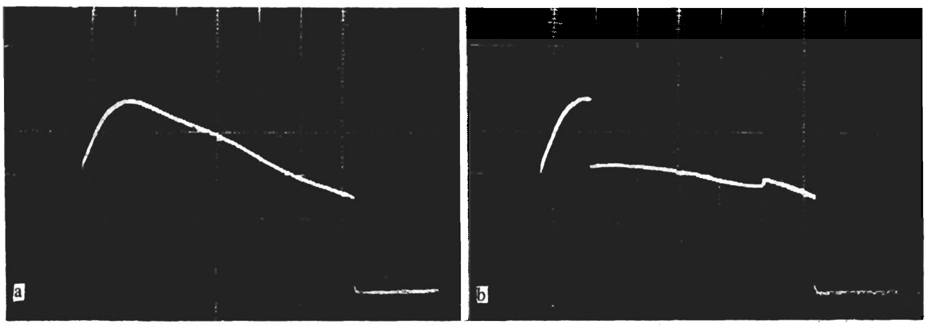

FIG. 2. Oscillograms of pulses: a—Pulse passing through resonator in the absence of beyond-threshold absorption in the sample. b—Pulse in the presence of such absorption.

the temperature interval 1.2–2.4°K. The temperature was determined by measuring the pressure of the saturated helium vapor with a McLeod manometer. The temperature was measured and maintained with accuracy not worse than 0.01°K.

EXPERIMENTAL RESULTS

By applying to the resonator with the sample a rectangular microwave pulse, we observed that when the amplitude of the microwave field exceeded a certain critical value h_{c1} a break appears on the pulse and corresponds to a sharp increase of the absorption in the sample. It is important to note that the size of the break (the power ΔP_1 absorbed by the sample does not tend to zero as $h/h_{c1} \to 1$, but to a certain finite value that depends on the static field and on the sample temperature, while the amplitude of the microwave field on the sample falls below the threshold value past the break.

With increasing h/h_{c1}, the time τ_1 from the start of the pulse to the break decreases. The results of these experiments, for different static fields and temperatures, show that $1/\tau_1$ is linearly connected with h (Fig. 3). At the minimum attainable excess $h/h_c = 1.02$, determined by the stability of our generator, we obtained $\tau_1 \approx 0.5$ msec.

For a more detailed study of the onset of absorption in the sample, we perfomed experiments with long (≈15 msec) pulses of special waveform (Fig. 2a). Whenever the maximum value of h exceed h_{c1}, the pulse passing through the resonator acquired the form shown in the oscillogram of Fig. 2b. It is seen from the oscillogram that not only the onset but also the vanishing of the absorption occurs jumpwise. The latter occurs at a field $h = h_{c2}$ at the sample.

Thus, the investigated phenomenon is characterized by two critical fields h_{c1} and h_{c2}. Figure 4 shows plots

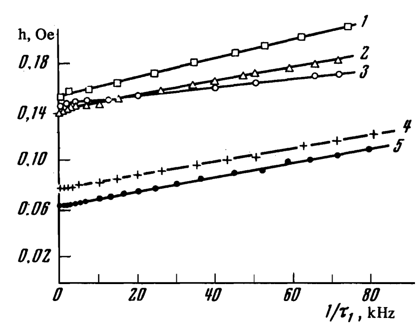

FIG. 3. Dependence of the reciprocal of the time τ_1 to the "break" on the microwave field h at the sample: 1—T = 1.98°K, H = 0.39 kOe; 2—T = 1.20°K, H = 0.39 kOe; 3—T = 1.54°K, H = 0.39 kOe, 4—T = 1.54°K, H = 2 kOe; 5—T = 1.20°K, H = 2 kOe.

of these fields against the static field H for several temperatures.

In[3,4] there were observed anomalies in the beyond-threshold susceptibility of $MnCO_3$ at ν_p = 36.0 GHz and in fields H_1 = 3.37 kOe and H_2 = 3.7 kOe, the former being apparently due to the presence of impurities in the crystal and the latter to magnon-phonon interaction. The investigations have shown that h_{c1} increases during the pulses in the field H_1, and both fields h_{c1} and h_{c2} increase in the field H_2, but the changes are small and lie beyond the accuracy of our experiment.

DISCUSSION OF RESULTS

If it is assumed that the number of parametrically excited magnons increases in accordance with formula (2), then the amplitude of the pulse passing through the resonator should decrease monotonically at $h > h_{c1}$. By determining the damping $\Delta\nu$ from the value of the threshold field, we can easily determine the time τ

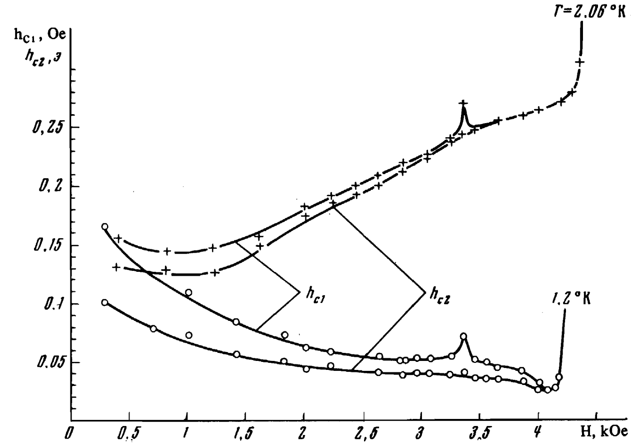

FIG. 4. Dependence of the critical fields h_{c1} and h_{c2} on the static field H.

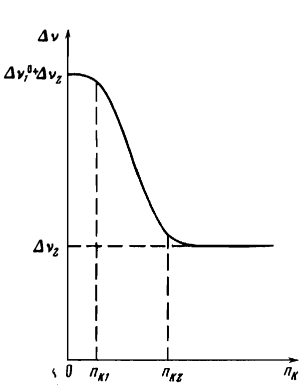

FIG. 5. Proposed dependence of the relaxation $\Delta\nu$ on the spin-wave density n_k.

when, at a definite excess above threshold, the number of spin waves increases enough to be able to register absorption in our experiment. It turns out that τ for $h/h_c = 1.02$ should be of the order of 20 msec. As shown by the experiment, the absorption increases in a different manner: the absorption in the system increases sharply at an instant $\tau_1 \ll \tau$, that depends on the excess above threshold, while the field at the sample falls below h_{c1} at $t > \tau_1$. This allows us to conclude that the growth of the number of magnons in the investigated antiferromagnet proceeds apparently in two stages: from the start of the pulse to the instant τ_1 the number of magnons increases in accord with the exponential law (2), but the energy absorption corresponding to this process cannot be registered in our experiment. This is followed by an avalanche-like growth of the number of magnons and, as follows from experiments on the determination of τ_1 as a function of the microwave field amplitude (Fig. 3), this avalanche sets in at a definite number of magnons.

The results of experiments with rectangular and non-rectangular pulses can be easily explained by assuming that the spin-wave relaxation consists of two parts: $\Delta\nu = \Delta\nu_1 + \Delta\nu_2$ where $\Delta\nu_2$ depends little on the spin-wave amplitude, and $\Delta\nu_1$ decreases rapidly with increasing n_k, from an initial value $\Delta\nu_1^0$ corresponding to $n_k = 0$ (Fig. 5).

When a field barely exceeding h_{c1} is applied to the sample, the number of magnons begins to grow in accord with (2). After a time τ_1 their number increases

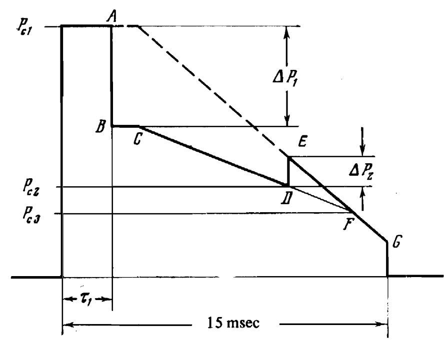

FIG. 6. Schematic representation of the non-rectangular pulse whose oscillogram is shown in Fig. 2b.

to n_{k1}, at which $\Delta\nu_1$ begins to be turned off. With decreasing $\Delta\nu_1$, the threshold field decreases, causing the rate of growth of the magnon number to increase, causing in turn a further decrease of $\Delta\nu_1$. An avalenche-like growth of n_k sets in and leads to a complete turning-off of $\Delta\nu_1$ within a time 10–20 μsec. A stationary magnon density is established in the system and is determined by the excess of the sample field above the critical value $h_{c3} \sim \Delta\nu_2$, and the spin system "forgets" the existence of $\Delta\nu_1$. If the microwave-field amplitude is now lowered (Fig. 2b, 6), then the equilibrium number of magnons also begins to decrease, as evidenced by the decrease in the power absorbed by the sample. When the magnon density becomes equal to n_{k2} ($h = h_{c2}$), the relaxation $\Delta\nu_1$ is turned on. An inverse avalanche is produced, viz., the smaller the number of spin waves the larger $\Delta\nu_1$, and the larger $\Delta\nu_1$ the faster the spin-wave damping. The relaxation $\Delta\nu_1$ is completely turned on, and since the field at the sample is now less than h_{c1}, the parametric excitation ceases and the spin-wave amplitude decreases to the thermal background value. From the data of our experiment we can calculate the relaxation $\Delta\nu_1^0$ and $\Delta\nu_2$, and also the magnon densities n_{k1} and n_{k2}. Figure 6 shows schematically the pulse whose oscillogram is shown in Fig. 2b. All the experimentally measured quantities needed for the calculation are marked on this figure.

The field h_{c3} was calculated from the value of the power at the point F, determined by the intersection of the continuation of the line CD with the line EG. In our experiments, the field h_{c3} differed from h_{c2} by not more than 10%. The relaxation frequencies $\Delta\nu_1^0$ and $\Delta\nu_2$ were calculated from h_{c3} and h_{c1} using the formulas

$$\Delta\nu_2 = h_{c3}(2H + H_D) / \gamma^2\nu_p,$$
$$\Delta\nu_1{}^0 = h_{c1}(2H + H_D) / \gamma^2\nu_p - \Delta\nu_2. \quad (3)$$

The results of these calculations are shown in Figs. 7 and 8.

The relaxation $\Delta\nu_1^0$ decreases rapidly with increasing magnetic field, and vanishes at $H = H_c$, corresponding to $k = 0$. In a field $H_1 = 3.37$ kOe, a resonant increase of this relaxation is observed. $\Delta\nu_1^0$ decreases with increasing temperature.

The relaxation $\Delta\nu_2$ varies linearly with the square of the magnetic field up to fields of 3.7 kOe, i.e., in this interval we have

$$\Delta\nu_2 = \Delta\nu_{20}(T) + \beta(T)H^2, \quad (4)$$

$\Delta\nu_{20}$ depends little on the sample temperature and equals $\approx$0.1 MHz.

If we calculate the magnon mean free path λ corresponding to $\Delta\nu_2 = 0.1$ MHz and $H = 0$ ($k \approx 6 \times 10^5$ cm^{-1}), then we obtain $\lambda \approx 1.5$ mm, which coincides approximately with the dimensions of our samples. This means apparently that $\Delta\nu_{20}$ is determined mainly by the scattering of the magnons by the crystal boundaries, i.e., by the relaxation $\Delta\nu_{lim} = L(2\pi d\nu/dk)^{-1}$. The dependence of $\Delta\nu_{lim}$ at $L = 2$ mm on the magnetic field, which is connected with the change of the magnon velocity, is shown dashed in Fig. 8.

The quantity β obeys the power law $\beta \sim T^{7.4}$ in the temperature interval 1.2–2.19°K. Such a high power can hardly have a physical meaning. It is more likely that β depends on some exponential functions of the temperature.

From the results of our experiments we can calculate the magnon densities n_{k_1} and n_{k_2} at which the relaxation $\Delta\nu_1$ is "turned on or off". The value of n_{k_1} was calculated from formula (2), where n_{k_0} was taken to be the thermal density

$$n_{k0} = \{\exp[h\nu / k_B T] - 1\}^{-1}.$$

n_{k_2} can be estimated from the power absorbed by the sample at the instant when the "turning on" of the relaxation begins (section DE in Fig. 6). The power absorbed in the sample can be calculated from the formula

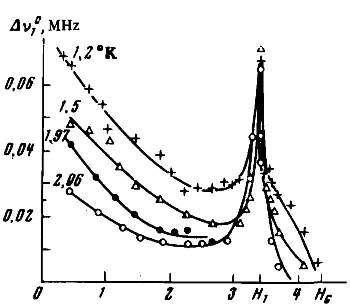

FIG. 7. Relaxation $\Delta\nu_1^0$ vs the magnetic field at various temperatures.

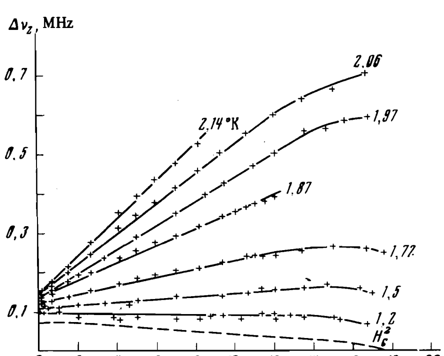

FIG. 8. Dependence of the relaxation $\Delta\nu_2$ on the square of the magnetic field at different temperatures. Dashed—dependence of $\Delta\nu_{lim}$ on H^2.

$$\Delta P_2 = {}^1/_2 h\nu_p N(k) \cdot 2\pi\Delta\nu_2, \quad (5)$$

where

$$N(k) = n_{k2} 4\pi k^2 V \Delta k / (2\pi)^3$$

is the number of parametrically excited magnons with wave vector **k**, and V is the volume of the sample.

The interval of the excited spin waves Δk was calculated from the condition

$$|{}^1/_2\nu_p - \nu_k| < {}^1/_2\Delta\nu_2[(h / h_{c3})^2 - 1]^{1/2}, \quad (6)$$

by starting from the value of $\Delta\nu_2$ and the known excess h/h_{c3} at the instant when the relaxation $\Delta\nu_1$ is "turned on."

The condition (6) was obtained by a method similar to that used in[7] from the known spin-wave dispersion law. The values of n_{k_1} and n_{k_2} calculated in this manner are given in the able.

T, °K	k·10^{-5}, cm^{-1}	H, kOe	n_{k0}	n_{k1}	$n_{k2} \cdot 10^{-5}$
1,98	7,37	0,39	1,82	150	2
1,54			1,35	130	1,5
1.2			0.96	100	1,2
1,98	6,0	2.0	1.82	—	1,5
1,54			1,35	780	1,0
1,2			0,96	770	0,7

The existence of a relaxation $\Delta\nu_1$ that becomes turned off and leads to a "hard" excitation of spin waves in antiferromagnets can be brought about by several mechanisms.

An analogous phenomenon was observed in yttrium iron garnet by Le Gall et al.[8], who proposed the following explanation: It was assumed that the relaxation was due mainly to a three-magnon process, viz., collision between magnons with k_1 and k_2 produces a magnon with $k_3 = k_1 + k_2$; the relaxation is determined by the densities n_{k_2} and n_{k_3}. At a small excess above threshold, n_{k_2} and n_{k_3} are equal to their thermal values $n_{k_{20}}$ and $n_{k_{30}}$, which determine the initial relaxation. With increasing n_{k_1}, the values of n_{k_2} and n_{k_3} change in such a way that the relaxation decreases. In our case, apparently, the principal role in the relaxation is played by a three-magnon process[6], and the described mechanism can take place.

It is also possible that the observed behavior of the relaxation is due to scattering of the magnons by the magnetic impurity ions. Favoring this assumption is the fact that $\Delta\nu_1^0$ increases with decreasing temperature, and indeed a peak is observed on the $\Delta\nu_1^0(h)$ plot in the field H_1, a peak interpreted in[3,4] as the consequence of a resonant interaction with an impurity mode.

We are deeply grateful to P. L. Kapitza for interest in the work. We sincerely thank A. S. Borovik-Romanov, and V. S. L'vov for a discussion of the results, and V. I. Ozhogin and A. Ya. Yakubovskiĭ for useful discussions.

[1]M. H. Seavey, Phys. Rev. Lett., 23, 132 (1969).
[2]L. A. Prozorova and A. S. Borovik-Romanov, ZhETF Pis. Red. 10, 316 (1969) [JETP Lett. 10, 201 (1969)].
[3]B. Ya. Kotyuzhanskiĭ and L. A. Prozorova, ibid. 13, 430 (1971) [13, 305 (1971)].

[4]B. Ya. Kotyuzhanskiĭ and L. A. Prozorova, Zh. Eksp. Teor. Fiz. 62, 2199 (1972) [Sov. Phys.-JETP 35, 1050 (1972)].
[5]H. Yamazaky, J. Phys. Soc. Japan, 29, 1383 (1970).
[6]V. I. Ozhogin, Zh. Eksp. Teor. Fiz. 58, 2079 (1970) [Sov. Phys.-JETP 31, 1121 (1970)].
[7]L. D. Landau and E. M. Lifshitz, Mekhanika (Mechanics), Nauka 1965, p. 103.[Addison-Wesley, 1969].
[8]H. Le Gall, B. Lemaire, and D. Sere, Solid State Somm., 5, 919 (1967).

Translated by J. G. Adashko
241