Heating and energy diffusion in a coupled electron–nuclear magnetic system of antiferromagnetic $CsMnF_3$ subjected to strong parametric excitation

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An investigation was made of the spectra of electromagnetic radiation emitted by antiferromagnetic $CsMnF_3$ at various stages in a parametric pulse exciting nuclear spin waves. It was found that the radiation appeared at the instantaneous NMR frequency. A threshold field was necessary to heat the magnetic system of the nuclei to a certain temperature. The heating time was 0.1–100 ms and the reciprocal heating time was proportional to the square of the amplitude of the alternating pump field. A kinetic instability in a system of parametrically excited nuclear spin waves is regarded as a possible mechanism of energy diffusion of magnons from a $(\omega_p/2, k)$ state to a state with $\omega_{\rm NMR}$, $k = 0$ (ω_p is the pump frequency).

Antiferromagnets with a weak crystalline anisotropy and containing Mn^{2+} ions exhibit a low-frequency branch in the spectrum of magnetic excitations associated with simultaneous oscillations of the antiferromagnetic and nuclear magnetic systems.[1] This branch had been investigated in detail experimentally[2-4] and its excitation spectrum can be described quite reliably by an expression obtained in Ref. 1:

$$\omega_k^2=\omega_n^2\left(1-\frac{2\gamma^2H_EH_N}{\Omega_{ek}^2}\right), \qquad (1)$$

where ω_n is the nuclear magnetic resonance (NMR) frequency in the hyperfine field of the electron system of the crystal; $\Delta_N^2 = 2\gamma^2 H_E H_N$ is the square of the antiferromagnetic resonance frequency in the effective hyperfine interaction field H_N, which is created by the nuclear magnetic system and acts on the electron antiferromagnetic sublattices; and Ω_{ek} is the low-frequency branch of the spectrum of excitations of the antiferromagnetic system derived allowing for the spatial dispersion and for Δ_N^2. The effect is easily observed if the quantities Δ_N^2 and Ω_{ek}^2 are comparable, so that it is necessary to ensure a weak crystalline anisotropy (and a weak external field $H_0 < 10$ kOe) as well as a strong hyperfine interaction (a high value of $\Delta_N{}^2$). In the case of $CsMnF_3$ investigated which we investigated the hyperfine field was $H_N = 9.15/T$ Oe, and the other parameters were $\Delta_N^2/\gamma^2 = (6.4/T)$ kOe^2 and $\omega_n = 666$ MHz.

The quantity $\Delta_N^2 \sim H_N$ is proportional to the average nuclear magnetization $\langle m\rangle$, which can be easily altered (reduced) by an hf field. Hence, the branch of normal excitations described by Eq. (1) is strongly nonlinear, i.e., its eigenfrequencies depend on the amplitude of the pump field. Such a dependence is observed when a branch at the NMR frequency ($k = 0$, homogeneous spatial oscillations),[5] and also in the case of parametric excitation of a branch with $k \neq 0$ by the method of parallel pumping at a frequency doubled relative to ω_k (Ref. 6).

If the branch described by Eq. (1) has a strong nonlinearity, depending on the power of the radiation at frequencies between ω_0 and ω_n, the system can be in two different states relative to $\langle m\rangle$. At low powers we have $\langle m\rangle \approx \langle m\rangle_T$, where $\langle m\rangle_T$ is the equilibrium nuclear magnetization (unsaturated state) at the selected temperature. The second (saturated) state is observed when the radiation power exceeds a certain threshold value and it corresponds approximately to the state of magnetization of the nuclear system $\langle m\rangle_\omega$ representing the solution of Eq. (1) for $\langle m\rangle$ when $\omega_0 = \omega$, which is the radiation frequency (or $\omega_p/2$ in the parallel pumping case). There is a fairly wide range of pump powers where both states can coexist, depending on the previous history of a sample (bistability region). We investigated earlier the process of transition from a saturated to an unsaturated state which is realized either on reduction of the pump power or in the absence of external power (see Ref. 5 and the bibliography cited there). In the present study we were able to obtain information on the process of transition from an unsaturated to a saturated state in a sample of a hexagonal antiferromagnet $CsMnF_3$ with an easy-plane anisotropy.

METHOD

We investigated the spectra of the radiation emitted by the sample during a parametric pulse which heated the nuclear system and we recorded the change in the NMR frequency under the influence of such a pulse. A sample of $CsMnF_3$ in the form of a plane-parallel plate of thickness ~ 1 mm was placed inside a three-turn coil used to detect the radiation generated in the sample. The coil in turn was located inside a helical resonator 5 mm in diameter and 10 mm long used in parametric excitation of nuclear spin waves. The axes of the coil and of the helical resonator were mutually perpendicular and the coil turns did not screen the sample from the pump field.

The power of electromagnetic radiation emitted by the sample and detected by the coil was passed on to a spectrum analyzer with an output in the form of a gating attachment, which made it possible to record the radiation spectra at various moments in time from the beginning of a parametric pulse. Our spectrum analyzer was not sensitive enough to detect the radiation from a sample, which made it necessary to place a preamplifier with a gain in excess of 40 dB in front of the detector.

The NMR investigation was made using a coaxial resonator consisting of a line of variable length, a coaxial cable, and a short-circuiting coil with a sample. The eigenfrequency of such a resonator was found to be of the order of

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100 MHz, but at higher resonator harmonics we could use a variable-length line and tune it to the required frequency. This coaxial resonator served as an absorption cell in a transmission-type NMR spectrometer. The NMR lines were observed in a static magnetic field and at a constant frequency. When a parametric pulse excited nuclear spin waves, the temperature of the nuclear spin system began to decrease and this, in accordance with Eq. (1), caused the NMR frequency to vary with time. When the NMR frequency coincided with the working band of the spectrometer, an NMR signal was observed.

RESULTS

Figure 1 shows the spectra of the radiation emitted by a sample at various moments in time from the beginning of a parametric pulse. As shown in Ref. 7, the emitted radiation was due to excitation of homogeneous precession (excitation of nuclear spin waves at $k \approx 0$) in a sample or some part of it.

Before arrival of a paramagnetic pulse the magnetization of the nuclear spin system was close to equilibrium at a given temperature and the corresponding NMR frequency was 540 MHz. This was precisely the frequency at which electromagnetic radiation was emitted by the investigated sample at the beginning of a parametric pulse. As the magnetic system temperature rose, the average magnetization of the nuclei decreased, corresponding to an increase in the NMR frequency of a sample and, as shown in Fig. 1, a shift of the radiation maximum toward higher frequencies.

Figure 2 shows the dependence of the position of the radiation maximum on the time elapsed from the beginning of a pump pulse, deduced from data similar to those shown in Fig. 1. During a parametric pump pulse the frequency of the emitted radiation increases monotonically and then (for a sufficiently high power in a pulse) intersects half the pump

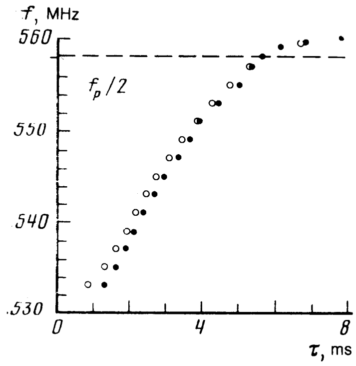

FIG. 2. Time dependence of the position of the radiation maximum (●); $\tau = 0$ represents the beginning of a parametric pulse; (○) is the time-dependent position of the NMR frequency; $f_p/2 = 558.3$ MHz.

frequency. Then it tends to some steady-state value corresponding to an overheated system of nuclear spins which are retained subsequently during a saturating pump pulse. In this state the NMR frequency is above the half pump frequency and the energy is delivered to the system either in the wing of the NMR line or due to excitation of magnetoelastic oscillations. In the parametric excitation case, nuclear spin waves creates pairs of magnons of frequency $\omega_p/2$ and with wave vectors k, $-k$, the values of which ($|k|$) are found by solving Eq. (1) on the assumption that $\omega_k = \omega_p/2$. We also observed the emission of radiation from a sample at a frequency different from $\omega_p/2$. Magnons created as a result of spin-spin relaxation began to "smear out" over the whole nuclear spin wave spectrum. The result of this energy diffusion was the appearance of magnons with $k = 0$ and a frequency ω' governed by the instantaneous temperature of the nuclear system. The appearance of such magnons excited homogeneous oscillations of the magnetization of the sample, which was the reason for the appearance of the recorded radiation. The width of the spectrum of the emitted radiation was governed by the width of the instantaneous inhomogeneously broadened NMR line. When the spectrum of the emitted magnons crossed the frequency $f_p/2$, the first excited magnons with $f = f_p/2$ and $k = 0$ began to appear, as manifested by a narrow line exhibited by the curves recorded 2.5–6 ms from the beginning of a pulse (Fig. 1). The intensity of this line was approximately 30 dB higher than the intensity of the secondary magnon line.

We also determined the time dependence of the position of the NMR line after application of a parametric pulse (Fig. 2). The dependence $\omega_{NMR}(t)$ was basically the same as the dependence of the position of the maximum in the emission spectrum. The difference between them was as follows: the radiation emitted by the specimen lagged in time relative to the position of the NMR line. This was due to the fact that excitation of homogeneous oscillations of the magnetization required some specific time to ensure energy diffusion of magnons to the $k = 0$ state.

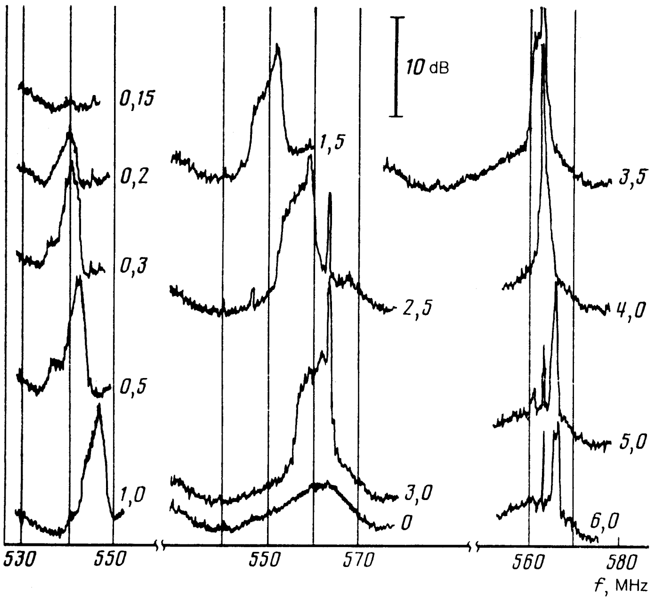

FIG. 1. Spectra of electromagnetic radiation emitted by a sample at various moments during a parametric pulse. The pump frequency was $f_p = 1126$ MHz, applied in the presence of a field $H = 3.1$ kOe at $T = 1.27$ K. The curve denoted by 0 is the zero-load curve of a spectrometer with an amplifier. The numbers alongside the curves give the time from the beginning of a pulse (in milliseconds).

Figure 3 shows the dependence of the reciprocal of the time corresponding to the position of the maximum of the radiation emitted at frequencies 528.5 and 535 MHz on the pump pulse power. The pump frequency was $f_p = 1099$ MHz and the frequency 528.5 MHz represented the position of the NMR line before arrival of the parametric pump pulse

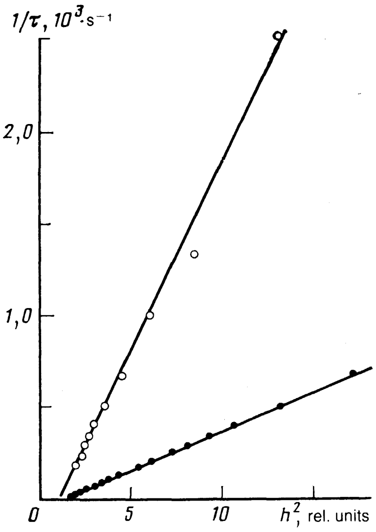

FIG. 3. Dependence of the reciprocal of the time corresponding to the position of the radiation maximum ($1/\tau$) on the relative power in the resonator: ○) frequency 528.5 MHz; ●) 535 MHz. Pump frequency f_p = 1099 MHz.

(equilibrium magnetization), whereas the frequency 535 MHz corresponded to an increase in the temperature of the nuclear system by ~0.1 K.

The time of observation of the radiation maximum corresponded to a very definite state of the magnetic system of nuclear spins, which depended on the total number of excited magnons. This number was proportional to the energy absorbed by the sample

$$E=\int_0^\tau h^2\chi''(t)\,dt\approx h^2\int_0^\tau \chi''(t)\,dt. \qquad (2)$$

Here, τ is the time at which the maximum was observed and $\chi''(t)$ is the imaginary part of the susceptibility of the sample. Since the magnetic field h in the resonator changed little with time (by a few percent), we could take h^2 outside the integral.

Assuming that $\chi''(t)$ depended only on the instantaneous state of the system (temperature), i.e., that the change in h in the resonator due to a change in the power reaching it corresponded to compression or dilatation of the function χ'' with time, we found that

$$E\propto h^2\tau=\text{const}, \qquad (3)$$

yielding the dependence $1/\tau \propto h^2$.

It is clear from Fig. 3 that there was a threshold field needed to heat the magnetic system of the nuclei to a certain temperature in the limit $\tau\to\infty$. For example, on increase in the temperature of a magnetic system of the nuclei by 0.1 K (corresponding to the frequency 535 MHz) the threshold field had to be about 20 dB higher than the critical field for parametric excitation of nuclear spin waves.

It should be pointed out that in this context the threshold field should be regarded as the field in the resonator and not the alternating field in the sample, since the phase mechanism which limits the number of spin waves makes the field in the sample, i.e., the sum of the external alternating field and the induction field of spin waves, equal to the critical field. The field in the resonator is $\approx$20 dB higher than the critical field of the parametric instability, which shows that the phase limitation mechanism is active and a large number of nuclear spin waves are excited.

DISCUSSION

One of the possible energy diffusion mechanisms is a kinetic instability in a system of parametrically excited spin waves. A kinetic instability proposed by Lavrinenko *et al.*[8,9] represents a four-magnon process in which-two parametric spin waves with wave vectors $k=-k'$ and of frequency $\omega_k=\omega_{k'}=\omega_p/2$ decay into two spin waves with the same total wave vectors and frequencies, but with $k_1\neq k_2$ and $\omega_{k_1}\neq\omega_{k_2}$. In the case of a ferrite the spectrum of electron spin waves represents a wide band which ensures that a kinetic instability can occur (when the laws of conservation of energy and momentum are obeyed). In the case of nuclear spin waves in an antiferromagnet the direct process of kinetic instability is most probably impossible in a system of parametrically excited magnons, because the dependence $\omega(k)$ is nonlinear and represents a narrow band in the (ω, k) plane.

The main mechanism of relaxation of parametrically excited nuclear spin waves is the scattering by paramagnetic fluctuations. This mechanism is analogous to the scattering of light in a turbid medium, i.e., when the magnon frequency is conserved in the scattering process. The scattering of parametrically excited nuclear spin waves creates a large number of secondary spin waves which have the same energy as those which are excited and have wave vectors with all possible directions. If the curvature of $\omega(k)$ is positive, which is true in the present case of nuclear spin waves, the conservation laws can always be satisfied. An elementary kinetic instability event involves one spin wave excited directly by parametric pumping (when the density is significantly higher) and a spin wave from a secondary system belonging to a group that satisfies the laws of conservation of energy and momentum.

Our results demonstrate that strong parallel pumping heats the nuclear system and creates radiation emitted at a frequency close to the homogeneous resonance frequency corresponding to the instantaneous temperature of the nuclear system. This means that secondary nuclear magnons with wave vector $k\approx 0$ are created. They may not be the only ones since in the $k\gg 1/d$ case (where d is the characteristic dimension of a sample) the dipole emission of an electromagnetic wave is not detected because the phase of the magnetization oscillations fluctuates rapidly over the volume of a sample i.e., we can assume that magnons are excited throughout the whole zone from $k=0$ to $k\approx 2k_p$ (where k_p is the wave vector of the primary parametric magnons).

The appearance of a kinetic instability corresponds to a steeper rise of the rate of relaxation of primary magnons. This should suppress the phase mechanism which limits the absorption of the energy from an hf magnetic field, so that this energy increases and the pump power is acquired effectively by the whole magnon energy band mentioned above, resulting in heating of the nuclear system. Govorkov and Tulin[11] observed the threshold process corresponding to heating of the nuclear system to a state when the NMR frequency ($k=0$) became comparable with half the pump frequency. The threshold field of this process can be identified with the critical field of a kinetic instability (curve 2 in Fig. 2—see Ref. 11).

CONCLUSIONS

The results of our investigations demonstrate that heating of the magnetic system of nuclei by parametric excitation of nuclear spin waves results in the emission of electromagnetic radiation from a sample at the instantaneous value of the NMR frequency. Magnons appear in a state with $\omega = \omega_{\rm NMR}$ and $k = 0$ because of their energy diffusion. A possible mechanism of this diffusion is a kinetic instability in a system of parametrically excited nuclear spin waves.

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