Above-threshold susceptibility in parametric excitation of magnons in antiferromagnetic $FeBO_3$

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The above-threshold susceptibility χ is investigated in parametric excitation of magnons in the easy-plane antiferromagnet $FeBO_3$. The magnons were excited by parallel pumping at a frequency $2\pi\cdot 35.6$ GHz. The experiments were performed at temperatures 1.6 and 2.2 K. It is shown that the above-threshold stationary state in the system of parametric magnons in $FeBO_3$ is determined by the positive nonlinear damping. The nonlinear damping parameter is found to be $a = 6\times 10^{-16}$ cm^3.

Much interest is attracted at present by the physics of nonlinear processes. Many characteristic properties of these processes can be described by nonlinear differential equations whose type does not depend on the concrete nature of the anharmonicity.

One of the most informative and simple experimental methods used now to study these phenomena is parametric excitation of magnons in magnetically ordered substances, and investigation of the produced stationary magnon state above the parametric-excitation threshold.

Investigations of the stationary state, which were reduced mainly to a determination of the dependence of the nonlinear high-frequency susceptibility $\chi = \chi' + i\chi''$ on the amplitude of the exciting alternating magnetic field $\mathbf{h}$ (defined in accord with $\mu_\parallel = \chi h$, where $\mu_\parallel$ is the magnetization component which is parallel to $\mathbf{h}$ and oscillates at the exciting-frequency field), were initiated in the middle Fifties by Damon[1] and by Bloembergen and Wang.[2] Dating back to the same time is the work by Suhl,[3] who was the first to investigate theoretically an interaction in a system of exciting magnons. He has shown that the amplitude of the homogeneous ferromagnetic resonane is limited by the parametric instability.

The principal experimental method of magnon excitation is at present that of parallel pumping, predicted for ferromagnets by Morgentaller[14] and by Schlömann, Green, and Milano,[15] and experimentally observed by the latter. In this case the microwave field $\mathbf{h}$ is applied parallel to the static field $\mathbf{H}$. At sufficiently large amplitude h (i.e., $h > h_c$, where h_c is the threshold parametric-excitation field), magnon pairs are produced in the field and have a frequency ω_k equal to half the pump frequency ω_p, as well as equal and opposite wave vectors $\mathbf{k}$ and $-\mathbf{k}$. The spin waves corresponding to these magnons can be represented in the form

$$\mu_{\mathbf{k}} \propto c_{\mathbf{k}} \cos(\omega_{\mathbf{k}} t - \mathbf{kr} + \varphi_{\mathbf{k}}),\quad \mu_{-\mathbf{k}} \propto c_{-\mathbf{k}} \cos(\omega_{-\mathbf{k}} t + \mathbf{kr} + \varphi_{-\mathbf{k}}), \tag{1}$$

where μ is the alternating component of the sample magnetization, $c_{\mathbf{k}}^2 = c_{-\mathbf{k}}^2 = n/2\hbar$, n is the magnon density, i.e., their number per unit volume of the sample, and $\hbar$ is Planck's constant.

The sum of these waves is the standing wave

$$\tilde{\mu}_{\mathbf{k}} \propto c_{\mathbf{k}} \cos(\omega_{\mathbf{k}} t + \psi_{\mathbf{k}}) \cos(\mathbf{kr} + \theta_{\mathbf{k}}). \tag{2}$$

The quantity $\psi_{\mathbf{k}} = (\varphi_{\mathbf{k}} + \varphi_{-\mathbf{k}})/2$ is called the combined phase of the pair and is determined by the phase and amplitude of the exciting microwave field h (the phase of the microwave field is assumed equal to zero). The microwave power P absorbed by the standing wave per unit sample volume, which determines the coupling of the pair with the pump field, is equal to[6]

$$P = 2hV_{\mathbf{k}}\omega_p c_{\mathbf{k}}^2 \sin\psi_{\mathbf{k}}, \tag{3}$$

where $V_{\mathbf{k}}$ is the coefficient of coupling with the pump, and has a maximum at $\psi_{\mathbf{k}} = \pi/2$. In this description, no conditions whatever are imposed on the difference phase $\theta_{\mathbf{k}}$.

According to prevailing opinions[6] the principal mechanisms that limit the number of parametric magnons are positive nonlinear damping, which was first pointed out in descriptions of parametric processes in magnets by Schlömann[7] and by Gottlieb and Suhl,[8] and the phase mechanism proposed by Zakharov, L'vov, and Starobinets.[9]

The first mechanism is connected with the increased relaxation parameter δ of the excited magnons as their density n is increased either by interaction of the magnons with one another[7,8] or by the rise of the effective temperature of other oscillations (e.g., spin and elastic) that interact with the parametric magnons.[7] In first-order approximation these interactions lead respectively to the following relations:

$$\delta = \delta_0(1 + an) \tag{4}$$

and

$$\delta = \delta_0(1 - 2n\hbar\delta_0/b)^{-1}, \tag{5}$$

where a is the nonlinear damping parameter, $b = cT_0/\omega_{\mathbf{k}}\tau$, c is the specific heat of the oscillations that interact with the parametric magnons, and T_0 and τ are their equilibrium temperature and relaxation time.

The second mechanism is connected with the dependence of the phase $\psi_{\mathbf{k}}$ of the pair on n: with increasing n the phase $\psi_{\mathbf{k}}$ decreases from the initial value ($\psi_{\mathbf{k}} = \pi/2$ at $h = h_c$), so that according to (3) the coupling of the pair with the exciting microwave field decreases. The mechanisms described lead to qualitatively different dependences of on the excess $\xi = h/h_c$ above threshold.

Both mechanisms are universal, i.e., they are always effective when the magnons are parametrically excited. Under certain conditions the positive nonlinear damping can be

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so strong that it determines the stationary state. This occurs, e.g., when the magnons are exited in the spectrum region in which their decay into other quasiparticles is allowed. In the case of a non-decay spectrum the principal role in the limitation of the number of magnons is played by the phase mechanism.

In the two aforementioned limiting cases, theory yields the following expressions for the above-threshold susceptibility χ. For the nonlinear damping mechanisms which lead to relations (4) and (5) we obtain respectively[7,8]

$$\chi'_{Na}=0,\quad \chi''_{Na}=\frac{2V_k\hbar}{ah_c}\frac{\xi-1}{\xi} \tag{6}$$

and

$$\chi'_{Nb}=0,\quad \chi''_{Nb}=\frac{1}{bh_c^2}\frac{\xi-1}{\xi^2}. \tag{7}$$

In the phase-limitation mechanism[6] we have

$$\chi_p'=\frac{2V_k^2}{S}\frac{\xi^2-1}{\xi^2},\quad \chi_p''=\frac{2V_k^2}{|S|}\frac{(\xi^2-1)^{1/2}}{\xi^2}, \tag{8}$$

where S is the four-magnon interaction constant. Experiments on the nature of the stationary states in ferromagnets are the subject of Refs. 10–16.

Experiments by Seavey[17] and by Prozorova and Borovik-Romanov[18] have shown that parametric excitation of magnons by parallel pumping takes place also in antiferromagnets. According to a theoretical study by Kolganov, L'vov, and Shirokov,[19] in antiferromagnets with easy-plane anisotropy (EPAF) as well as in ferromagnets, phase correlation of the waves takes place in the pairs and is responsible for the phase mechanism of the limitation. Therefore the stationary state in EPAF is determined by the two aforementioned limitation mechanisms.

Up to now, the stationary state was investigated in EPAF with nondecay spectra, $MnCO_3$ and $CsMnF_3$ (Refs. 20 and 21), in which the limitation is by the phase mechanism. For such antiferromagnets we have according to Ref. 22

$$V_k=\gamma^2(2H+H_D)/2\omega_p \tag{9}$$

and according to Refs. 19 and 23

$$S=-\gamma^4(4H+H_D)HH_E/2\omega_p^2M_0, \tag{10}$$

where $\gamma=2\pi\cdot 2.8$ MHz/Oe is the magnetomechanical ratio, H_D is the Dzyaloshinskii field, H_E is the exchange field, and M_0 is the sublattice magnetization. This yields[19] in accord with (8)

$$\chi_p=\chi_0\left[1+\frac{(2H+H_D)^2}{H(4H+H_D)}\frac{1-\xi^2+i(\xi^2-1)^{1/2}}{\xi^2}\right], \tag{11}$$

where χ_0 is the linear susceptibility, which coincides far from resonance with the static suceptibility $\chi_\perp=M_0/H_E$. Experiments 20 have confirmed these relations.

It can be seen from (11) that in weak ferromagnets ($H_D\neq 0$) and in weak fields the phase mechanism of limitation becomes ineffective ($\chi_p\to\infty$ as $H\to 0$). We note that a more rigorous theory that takes into account dipole-dipole interaction and two-magnon scattering by random defects[23] does not alter qualitatively the $\chi_p(\xi)$ dependence; in particular, it does not eliminate the divergences in weak fields.

We deemed it of interest to investigate the mechanism that limits the number of parametric magnons under these conditions. Our purpose here was therefore to investigate the above-threshold susceptibility of the weak feromagnet $FeBO_3$, which has a large $H_D=108$ kOe.

The magnetic properties of $FeBO_3$ have been thoroughly investigated. The basic magnetic parameters and a survey of the literature on this substance are given in Ref. 24, which reports also an investigation of parametric excitation of magnons in $FeBO_3$ by parallel pumping.

The study of the kinetics of parametric excitation of magnons in $FeBO_3$ (Ref. 24) has shown that at $T\lesssim 4.2$ K the process has a hard character. This means that it is characterized by two threshold fields h_{c1} and h_{c2} (with $h_{c1}>h_{c2}$): the excitation sets in at $h\geqslant h_{c1}$ and ceases at $h\leqslant h_{c2}$. It is assumed that the cause of this phenomenon is that relaxation of some part of the parametric magnons ceases when their number is increased above the thermal level (negative nonlinear damping). In this case the significant value for the stationary above-threshold field is apparently h_{c2}, and we shall substitute h_{c2} for h_c in all the expressions that follow.

EXPERIMENT

The magnons were parametrically excited by a direct-amplification microwave spectrometer by the procedure described in Ref. 25. A single-crystal $FeBO_3$ sample measuring $2\times 2\times 0.5$ mm in a small cigarette-paper container was secured to the bottom of a cylindrical cavity having a $Q\sim 10^4$ in the antinode of the H_{012} mode of the magnetic field. The fields **h** and **H** were in the basal crystal plane in which the direction of **H** could be varied during the experiment.

To permit measurement of the susceptibility in the field interval $h_{c2}<h<h_{c1}$ the magnetron microwave oscillator at a frequency $\omega_p=2\pi\cdot 35.6$ GHz and power 10 W operate in the regime of long (~ 1 msec) oblique pulses whose reptition frequency 50 Hz was produced by modulating the anode voltage. During the time of action of the microwave pulse the field h at the sample was gradually decreased. The change of the pump frequency can be neglected here, since it amounted during the time of action of the pulse to $\approx 2\pi\cdot 3$ MHz, and simple calculation shows that within the magnon relaxation time $\tau=\delta^{-1}\lesssim 0.2$ μsec the relative frequency change $\Delta\omega_p$ was less than $2\pi\cdot 1.5$ kHz, i.e., much less than δ^{-1}.

The measurements were made at 1.6 and 2.2 K. To improve the heat transfer from the sample, the cavity was filled with superfluid helium. The temperature was determined from the helium saturated-vapor pressure accurate to within ± 0.05 K. The microwave power imput to the cavity was measured with a thermistor bolometer. From this power and from the cavity parameters we determined the field h at the sample with absolute accuracy $\approx 20\%$ and relative accuracy $\approx 5\%$.

The above-threshold susceptibility $\chi=\chi'+i\chi''$ was determined from the change of the cavity resonance-characteristic change following the parametric excitation of the magnons in the sample:

$$\chi' = \chi_0 + \frac{1}{2\pi\sigma}\frac{\Delta\omega}{\omega_0}, \qquad (12)$$

$$\chi'' = \frac{1}{4\pi\sigma Q_H}\left[\left(\frac{p_0}{p}\right)^{1/2} - 1\right], \qquad (13)$$

where

$$\sigma = \int_{V_{samp}} h^2\,dV \Big/ \int_{V_{cav}} h^2\,dV$$

is the cavity filling factor, V_{samp} and V_{cav} are the volumes of the sample and cavity, χ_0 is the linear susceptibility ($\chi_0 \approx \chi_\perp = 1.6\cdot10^{-4}$ in our case), ω_0 and Q_H are the natural frequency and the loaded Q of the cavity with the sample $\Delta\omega = \omega - \omega_0$ is the shift of its frequency following the magnon excitation, and P and P_0 are the values of the power passing through the cavity in the presence and absence of excitation, respectively. The ratio P_0/P was measured, accurate to $\approx 10\%$, with a precision polarization attenuator placed ahead of the detector.

EXPERIMENTAL RESULTS

To describe the results of the investigation of the above-threshold susceptibility it is necessary first to measure the threshold field h_{c2}. Its values were determined by extrapolating to zero the dependence of the microwave power $P(h)$ absorbed by the sample above the excitation threshold on the field amplitude. Figure 1 shows a plot of $h_{c2}(H)$ measured at $T = 2.2$ K in a field **H** directed perpendicular in the crystal basal plane to the binary axis. In a field **H** directed along this axis the value of h_{c2} was somewhat higher, by not more than 20%. When the temperature was lowered to 1.6 K the field h_{c2} was decreased by approximately 1.5 times.

The values of χ'' calculated from (13) turned out to depend strongly on the magnitude and direction of the magnetic field **H** in the basal plane of the crystal. Figures 2a and 2b show plots of χ'' vs $(\xi - 1)/\xi$ for various values of H and for **H** directed parallel and perpendicular to the binary axis, respectively. The values of χ'' at a given $(\xi - 1)/\xi$ remained constant when the temperature was varied from 1.6 to 2.2 K.

Within the accuracy of our experiment, we observed no shift of the cavity resonant frequency above the threshold of the parametric excitation, all the way to $(h/h_{c2})^2 = 10$. This

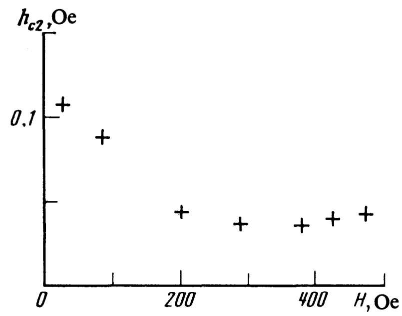

FIG. 1. Dependence of the threshold field h_{c2} on the strength of the static field **H** perpendicular to the binary axis, at $T = 2.2$ K.

means that $\chi' < 10^{-3}$ at all values of H at which magnon excitation was observed.

It can be seen from Fig. 2 that χ'' is directly proportional to $(\xi - 1)/\xi$. When **H** is perpendicular to the binary axis the proportionality coefficient increases with H in the interval $20 < H < 400$ Oe. The change with H of the slope of χ'' as a function of $(\xi - 1)/\xi$ when **H** is directed along the binary axis is smaller and does not exceed the experimental error.

DISCUSSION OF RESULTS

It follows from our results that in $FeBO_3$, in contrast to the previously investigated antiferromagnets $MnCO_3$ and $CsMnF_3$, the stationary state is determined not by the phase mechanism of limitation, but by the nonlinear positive damping (4). This is indicated by the absence of an appreciable real part χ' of the above-threshold susceptibility, which is at any rate less than $0.2\chi''$, whereas in the phase mechanism, according to Eq. (11), $\chi'_{max} = \chi''_{max}$ and the character of the dependence of χ'' on ξ agrees with (6).

According to (6) one can determine from the experimental data the positive nonlinear-damping parameter a. Within the limit of errors this parameter is independent of the magnetic field intensity H in the interval $20 < H < 480$ Oe and its temperature variation is such that $ah_{c2} = \text{const}$, i.e., a increases with decreasing temperature. At $T = 2.2$ K in a field **H** perpendicular to the binary axis we have $a = 6\times10^{-16}$ cm$^3 \pm 50\%$. In a field H directed along the binary axis the parameter a is noticably larger, e.g., by more than three times at $H = 380$ Oe.

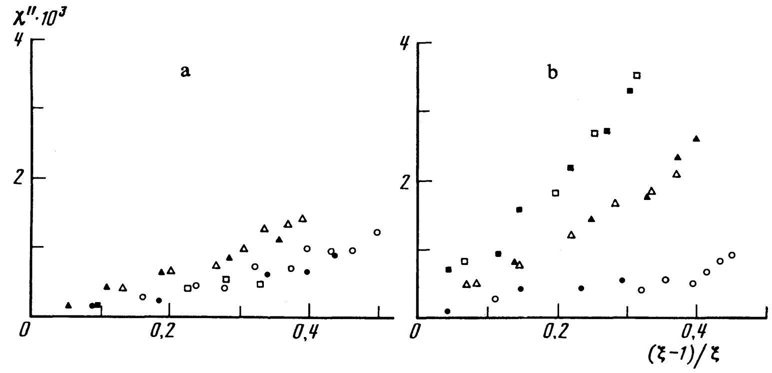

FIG. 2. Dependence of the imaginary part χ'' of the above-threshold susceptibility on $(\xi - 1)/\xi$ in a field **H** directed along the binary axis (a) and perpendicular to it (b): ○—$H = 25$ Oe, △—200, □—380; dark symbols—$T = 2.2$ K, light—1.6 K.

As already indicated, a fundamental role is played in the formation of positive nonlinear damping by three-particle decays (if they are possible) of parametric magnons. In $FeBO_3$ the energy and quasimomentum conservation laws allow the decay of a parametric magnon into a phonon and a magnon or into two phonons, since the maximum propagation velocity of the magnons in this substance exceeds that of sound. It is also known that these processes make a decisive contribution to the relaxation of parametric magnons.[24] It appears that the same processes determine also the positive nonlinear damping. One can verify this assumption and determine which of the aforementioned processes predominates in the formation of the nonlinear damping by calculating the contribution made to the parameter a by each of the indicated processes.

If this assumption is valid, the observed dependences of χ'' and of the parameter a on the direction of **H** in the basal plane of the crystal can be atributed to anisotropy of the magnetoelastic interaction.

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