Nonlinear properties of ferrites at low temperatures

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The imaginary part of the nonlinear susceptibility for parallel pumping of spin-wave instability has been investigated over the temperature range 5–300°K; self-modulation of the magnetization and the gap between the thresholds for appearance of instability and of self-modulation have also been investigated. The specimens studied were single-crystal spheres of yttrium-iron garnet. The pumping frequency was 9370 MHz. At low temperatures, substantial differences were detected in the nonlinear properties of the ferrites, as compared with the properties measured at room temperature. These differences consist in an increase of the role of nonlinear dissipative mechanisms that limit the amplitude of spin waves, an abrupt decrease of the self-modulation amplitude, and the disappearance of processes of hard excitation of spin waves.

In a ferromagnetic crystal located in a constant magnetic field H and, parallel to it, a microwave pumping field h of frequency ω, at a pumping power P equal to the threshold value P_H (which depends on H), there occurs parametric excitation of spin waves with frequency $\omega_k = \omega/2$. The value of P_H and the corresponding value of the threshold field h_H are found by means of the linear theory of parametric excitation of spin waves[1].

The nonlinear properties of ferrites that occur at $P > P_H$ or $h > h_H$ (such as the imaginary and real parts χ'' and χ' of the nonlinear susceptibility, the self-modulation amplitude, etc.) must be described by means of the nonlinear theory, which is at present undergoing intensive development[2,3]. But there is still no unified view on the nature of the mechanism of limitation of the amplitude of spin waves after the threshold of excitation[4], and knowledge of this mechanism seems an important condition for the construction of a correct theory of parametric excitation of spin waves in ferrites. The experimental results likewise give no unambiguous answer[5].

Here it should be noted that the majority of the experiments have been conducted on monocrystals of yttrium iron garnet (YIG) at room temperature; only the self-modulation of parametric resonance has been investigated at nitrogen temperatures[6]. Measurement of the nonlinear properties with various parameters is very desirable and can be accomplished by extension of the class of crystals investigated or by investigation of YIG single crystals at various temperatures, since it is known that the properties of YIG depend importantly on temperature; for example, the spin-wave ling ΔH_k near the helium temperature is more than an order of magnitude narrower than at room temperature[7].

In the present work, a detailed investigation is made of the value of the imaginary part of the nonlinear susceptibility in parallel pumping of monocrystalline spheres of YIG, over the temperature range 5 to 300 K. At these temperatures a study is made of the amplitude of self-modulation, its frequency, and the threshold for its onset. The experiment was conducted in the 3-cm range of wavelengths.

EXPERIMENT

1. The investigation was conducted on the usual experimental apparatus for observation of parametric excitation of spin waves, like that described earlier[8]. Here we mention only that the pumping frequency was 9370 MHz, the duration of the pumping pulse 200 μsec, and the pulse repetition frequency 50 Hz. For measurement of the temperature of the ferrite specimen, the resonator with the ferrite was placed inside the column of a cryostat filled with helium vapor. The specimen temperature could be set over the range 4.2 to 300 K with a constancy of $\pm$ 0.02 K, while the absolute value of the temperature was measured with an accuracy of $\pm$ 0.25 K.

2. Figure 1 shows the experimental dependence of χ'' on the value of the external magnetic field H for two temperatures, 300 and 5 K. The measurements of susceptibility were made at a constant excess over the pumping threshold level, P/P_H = 2 dB. The figure corresponds to the case of orientation of the external constant magnetic field H along the easy axis [111] of the crystal.

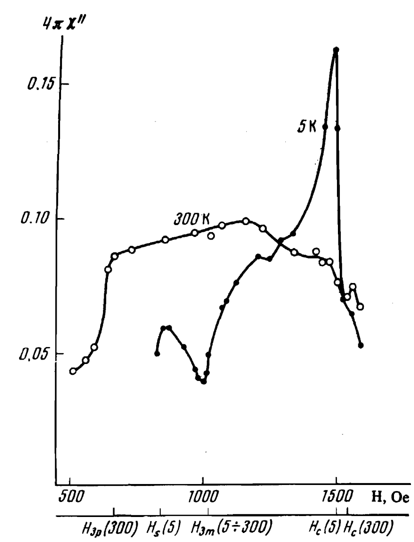

FIG. 1. Dependence of the imaginary part of the nonlinear susceptibility on the value of the external constant magnetic field, for a YIG sphere of diameter 2.25 mm. The numbers on the curves give the measurement temperature. Excess of pumping over the threshold level, P/P_H = 2 dB. Specimen magnetized along the easy axis [111]. On the horizontal axis are plotted the values of the fields for three-magnon splitting H_{3p}, saturation H_s, three-magnon fusion H_{3m}, and minimum threshold of spin-wave instability H_c, for the respective specimen temperatures.

The room-temperature dependence $\chi''(H)$ differs to no significant degree from that found earlier[8]. According to this dependence, the value of χ'' at room temperature, for orientation of H along the [111] axis, is approximately constant over the range of fields from H_{3p} to H_c, where H_{3p} is the upper bound of constant magnetic fields at which the process of three-magnon splitting is possible[8], and where H_c is the value of the constant magnetic field at which spin waves with wave vector k = 0 are excited; the threshold of spin-wave instability is a minimum here[1]. For the case shown in Fig. 1, H_{3p}(300 K) = 660 Oe, H_c(300 K) = 1540 Oe. Of course, like the other characteristic fields (such as the saturation field H_s, the three-magnon fusion field H_{3m} at which it becomes possible for two parametric magnons to fuse into a single resultant magnon[9], etc.), H_{3p} and H_c depend on temperature, since with change of temperature the magnetization and crystallographic anisotropy of the ferrite change; therefore in Fig. 1, along with the field designations on the horizontal axis, the temperature is shown in parentheses.

The decrease of the susceptibility at $H < H_{3p}$ has been explained theoretically by L'vov[10], and the decrease at $H > H_c$ may be due to the inapplicability for $k \to 0$ of the plane spin-wave model that is usually used in the theory. The slight increase of χ'' in fields $H \lesssim 1340$ Oe is explained by the mechanism of hard excitation of spin waves[10, 11].

Entirely different is the behavior of $\chi''(H)$ measured at temperature 5 K. Here there is no question of any constancy of the susceptibility at fields $H < H_c$, as for T = 300 K.

We note an interesting peculiarity. At temperature 5 K and field H parallel to the direction [111], three-magnon splitting processes are impossible in a saturated monocrystalline YIG sphere; as a result, the decrease of χ'' in fields $H \lesssim 850$ Oe is due to transition of the ferrite into an unsaturated state, and not to three-magnon splitting processes. Furthermore, for orientation of H along the [111] axis the three-magnon fusion field H_{3m} is practically independent of temperature.

3. Figure 2 shows the evolution of the $\chi''(H)$ curves in their dependence on temperature of the ferrite, which this time is oriented with its hard axis [100] along the constant magnetic field. The curve for room temperature differs here from the analogous curve in Fig. 1. In the first place, for magnetization of the crystal along the hard axis two mechanisms of hard excitation of spin waves are acting, rather than one. The first mechanism, independently of the crystal orientation, acts in fields below $H' \approx H_c - 200$ Oe (according to[10]; H' = 1480 Oe for H ∥ [100] and H' = 1340 Oe for H ∥ [111]). In both cases this mechanism leads to a slight increase of the susceptibility with decrease of the field below the value H'. An additional mechanism of hard excitation of spin waves, discovered earlier[8] and similar to that investigated in[12], does not act for magnetization of the crystal along the easy axis; but for a crystal magnetized along the hard axis, it leads to an abrupt increase of χ'' in fields $H \lesssim 1250$ Oe, which is clearly evident in Fig. 2.

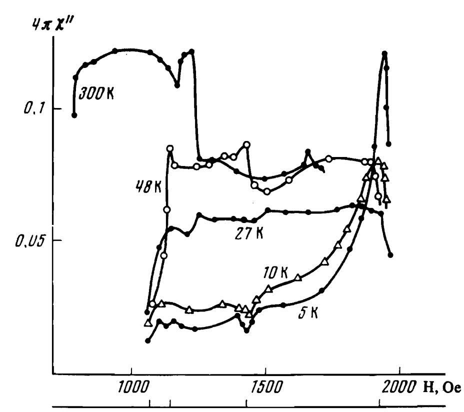

FIG. 2. Same as in Fig. 1, for magnetization of the ferrite along the hard axis [100]. The values of the fields H_s, H_{3p}, H_{3m}, and H_c on the horizontal axis correspond to specimen temperature 5 K.

A second peculiarity of the $\chi''(H)$ curve at room temperature, for magnetization of the crystal along the hard axis, is a significant decrease of the value of χ'' near the three-magnon fusion field H_{3m}(300 K) = 1150 Oe. We note that for a crystal magnetized along the hard axis, the value of H_{3m} depends significantly on temperature. As is evident from Fig. 2, H_{3m}(5 K) = 1435 Oe. The behavior of the curves for 5 K in Figs. 1 and 2 is in principle the same, although there is a slight anisotropy of the susceptibility with respect to orientation of the specimen in the magnetic field: χ'' is larger for magnetization of the crystal along the [111] axis than for magnetization along [100]. Clearly evident on both curves is the three-magnon fusion process, which shows up in a definite range of fields. The field interval and the relative decrease of χ'' are larger for orientation of the crystal along the easy axis, although the absolute value of χ'' remains smaller for magnetization along the [100] axis.

As is seen from Fig. 2, with increase of temperature there is a decrease of the difference between the maximum and minimum values of the susceptibility. Whereas for 5 K the susceptibility at $H = H_c$ exceeds by more than an order of magnitude the susceptibility at $H = H_{3m}$, at 10 K the ratio of the susceptibilities is close to 4. At a certain temperature of the ferrite, which we shall hereafter call the transition temperature T_t, we again have a situation in which χ'' is approximately constant over the field range from H_{3p} to H_c. For the orientation H ∥ [100], the temperature $T_t \approx 27$ K; for H ∥ [111], $T_t \approx 18$ K. For temperatures $T > T_t$ the functions $\chi''(H)$ are to a significant degree identical, as is seen by comparison of the curves in Fig. 2 taken at 48 and at 300 K; it should be noted, however, that with increase of temperature the jumps of susceptibility because of the influence of the additional mechanism of hard excitation of spin waves and the three-magnon fusion mechanism become larger. For magnetization of the crystal along the easy axis there are several peculiarities.

As was mentioned above, at room temperature, for such magnetization, there is no appreciable effect of the additional mechanism of hard excitation of spin waves (for H ∥ [100] there is a significant jump of the susceptibility for $H \lesssim 1250$ Oe). But at low temperatures (down to nitrogen), there is such an effect, and it leads to an abrupt increase of χ'' in constant magnetic fields a few tens of oersteds larger than H_{3m}.

4. At the transition temperature T_t there is a change not only in the character of the $\chi''(H)$ dependence, but also in the shape of the pulse reflected from the resonator containing the ferrite. Whereas for $T < T_t$ the de-

cay of the pulse has an exponential form, for $T > T_t$ it acquires a complicated form; in the simplest case, it is the sum of two exponentials shifted with respect to time[13]. This is evidence of the fact that the processes of hard excitation of spin waves begin to act only at a temperature $T \gtrsim T_t$.

5. At room temperature there is appreciable anisotropy of the excitation threshold and of the amplitude of self-modulation of the magnetization[2]. For magnetization of the crystal along the hard axis there occur only "weak" (in the terminology of Zautkin and Starobinets[14]) spatially inhomogeneous oscillations of the magnetization, whereas for magnetization along the easy axis there are "strong" homogeneous oscillations.

This picture persists for all $T > T_t$. For $T \lesssim T_t$, the anisotropy decreases abruptly because of suppression of the "strong" spatially homogeneous oscillations that occur for magnetization of the crystal along the easy axis. For such magnetization, for $T < T_t$ the amplitude of self-modulation is at least two orders of magnitude smaller than at room temperature; furthermore, a large gap appears between the threshold for excitation of parametric instability and the threshold (3 to 8 dB) for onset of self-modulation (at room temperature the gap amounts to several tenths of a decibel). There is also a change in the minimum frequency for self-modulation: as a rule, it increases with decrease of temperature, reaching at 5 K values of 250 to 350 kHz, whereas at room temperature the minimum frequency for self-modulation does not exceed 50 kHz.

6. Figure 3 shows the change of the nonlinear susceptibility χ'' (at 5 K), for longitudinal pumping, with increase of the amplitude of the field in the resonator, for two different values of the constant magnetic field: H_c and H_{3m}. Saturation sets in appreciably earlier in field H_c than in field H_{3m}. For specimen temperature 300 K, at fields $H_{3p} \leq H < H_c$ only dependences of the type of curve 1 in Fig. 3 are observed; dependences like curve 2 can be observed in fields $H < H_{3p}$, where three-magnon splitting of spin waves occurs.

7. It can be seen from Fig. 3 that the maximum values χ''_{max} at fields H_c and H_{3m} differ from each other considerably less than do the susceptibilities measured at constant excess over the threshold; for magnetization of the crystal along the hard axis this difference amounts to several tens of percent, while for magnetization along the easy axis it is less than the accuracy of measurement of χ'', which is 10%. On change of temperature from 5 K to room temperature, $4\pi\chi''_{max}$ varies within the limits 0.24 to 0.18 (for H ∥ [111]) or 0.16 to 0.18 (for H ∥ [100]). Thus the maximum susceptibility in longitudinal pumping depends very little on temperature and on the constant magnetic field.

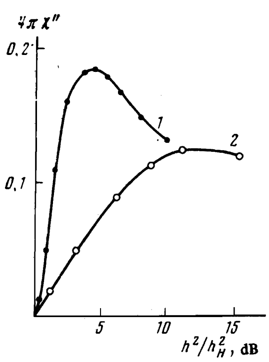

FIG. 3. Dependence of the imaginary part of the nonlinear susceptibility on the excess of the pumping field over the threshold level, for two values of the constant magnetic field: $H = H_c$ (curve 1) and $H = H_{3m}$ (curve 2). Specimen, a YIG sphere of diameter 2.25 mm. Specimen temperature 5 K.

DISCUSSION OF RESULTS

1. The most important feature of the $\chi''(H)$ curves (Figs. 1 and 2) is the presence of an essential difference between the curves obtained at different temperatures of the ferrite specimen. Whereas for $T > T_t$ the susceptibility is practically independent of the constant magnetic field, in the case of the reverse inequality there is a strong dependence of the susceptibility on H. As has already been mentioned the theories of the post-threshold behavior of ferrites[2, 3] explain fairly well the value and behavior of χ'' at room temperature, and precisely for this reason it is impossible by means of them, without some additional assumptions, to explain the behavior of the essentially different $\chi''(H)$ curves at $T < T_t$. In this case the curves are closest to the theory of Gottlieb and Suhl[9], based on a dissipative mechanism for limitation of the spin waves, although there are appreciable quantitative discrepancies in the relation between the susceptibilities measured at different fields. According to Gottlieb and Suhl, χ'' at H_c should be 3 to 4 orders of magnitude larger than at H_{3m}, but experimentally the difference amounts to only one order of magnitude.

One of the reasons for this discrepancy is probably the fact that Gottlieb and Suhl did not take account of the four-magnon phase mechanism[3], which is certainly much more probable than the five-magnon dissipative processes introduced in[9] for the calculation of the susceptibility near H_c. The presence of a minimum of χ'' for $H = H_{3m}$ indicates the occurrence of the three-magnon fusion process, whose probability disappears for spin waves with polar angle $\theta_k = \pi/2$, since the corresponding terms of the Hamiltonian vanish.

If we suppose that in parallel pumping of spin-wave instability, at least for small excesses above the threshold, there are only spin waves with $\theta_k = \pi/2$, then the presence of such a minimum can be explained as follows. It was shown in[12] that together with parametric magnons, there exist in the crystal so-called "intermediary" magnons, formed as a result of interaction of parametric magnons with the lattice and with thermal spin waves. The number of "intermediaries" is 5 to 8 times larger than the number of parametric magnons and, like the latter, increases with the pumping power. Hence it is clear that a mechanism of nonlinear attenuation is possible not only in the interaction of parametric magnons with parametric, but also in the interaction of parametric magnons with intermediary, for which θ_k may be different from $\pi/2$.

Here it should be stated that the mechanism considered may prove important not only at field H_{3m} but also at other fields, in particular at $H > H_{3m}$, since in principle it has no limitations with respect to the value of the constant magnetic field, although its probability must decrease with approach toward H_c.

Still another difference of the experimental curves from the theory of Gottlieb and Suhl is the fact that for $H \lesssim H_{3m}$ the susceptibility again increases, almost to the previous level. This difference may be due to the fact that the parametric spin waves do not have a uniform probability distribution with respect to the azimuthal angle φ_k but are concentrated near definite directions in the equatorial plane.

The fact that at 5 K and $H = H_{3m}$ a three-magnon fusion process is occurring, and not any four-magnon interaction (phase or dissipative), is supported by Fig. 3, where Curve 2 is very close to the theoretical $\chi''(h)$ curve for the three-magnon process[9]. Curve 1, on the contrary, which corresponds to the value $H = H_c$, is evidence of an appreciable contribution from four-magnon processes.

2. The data on the maximum susceptibility (χ''_{max} is approximately constant over the whole range of fields) speak in favor of the fact that at large excesses of the pumping over the threshold level (5 dB or more), the four-magnon phase mechanism exerts the dominant influence on the value of the nonlinear susceptibility χ'' in parallel pumping.

Summarizing what has been said, we may conclude that at $T < T_t$, for small excesses over the threshold field, the susceptibility in various ranges of constant magnetic field is determined by three- or four-magnon processes. At large excesses, the primary role is played by the four-magnon phase mechanism. This behavior of χ'' is quite natural, since for $h/h_H \rightarrow 1$ the three-magnon interaction, as a lower-order process, will always dominate over a four-magnon process, even if the latter has a larger coefficient in the Hamiltonian. At large excesses, the situation changes to the reverse. A certain discrepancy with the theory[3] must be noted in the quantitative determination of the value of χ''_{max}: the theory gives a value about two times larger than experiment. Furthermore, the theoretical variation of χ'' with the temperature of the ferrite is greater than the experimental.

3. An important difference of the experiments at $T < T_t$ is the absence of strong self-oscillations and the large gap between the threshold for excitation of spin waves and the threshold for excitation of self-oscillations. According to the existing theory of self-oscillations[2,15], this fact is unexpected, since on decrease of temperature no change occurs in the criteria for stability of collective oscillations of spin waves[15]. It may be supposed, however, that with diminution of temperature there is an increase in the role of two-magnon scattering of spin waves, which determines the gap between the instability threshold and the self-modulation threshold[16]. Here the increase in the role of two-magnon scattering occurs not because of an increase of its probability, but because of a strong decrease of the temperature-dependent probability of many-particle processes of relaxation of spin waves. In other words, against the background of a smaller value of the line-width ΔH_k of the spin waves, the influence of weak two-magnon processes will be noticeable. To test this assertion, we measured the value of ΔH_k at 5 K on a single ferrite specimen subjected to polishing of the surface by a variable abrasive paste with grain dimensions 0.5 to 20 μm. As is well known, at room temperature a different state of the surface does not lead to an appreciable change of the value of ΔH_k. In our experiments such a change was detected. For example, for spin waves with $k = 3 \cdot 10^5$ cm^{-1}, ΔH_k increased with coarsening of the surface from 0.045 to 0.06 Oe. The change in ΔH_k exceeded the error of measurement, which was0.1 ΔH_k. Consequently, at low temperatures two-magnon scattering can make a noticeable contribution to the relaxation processes and can, according to the theory[15,16], promote an increase of the threshold for occurrence of self-oscillations. It has also been established experimentally that two-magnon scattering affects the value of χ'', chiefly by decreasing the jump in the susceptibility near H_c.

4. Processes of hard excitation of spin waves become noticeable only at temperatures $T > T_t$. At room temperature, for small excesses over the instability threshold, such processes completely determine the shape of the susceptibility curve $\chi''(H)$[1)]. Absence of processes of hard excitation at low temperatures is not contained within the framework of L'vov's theory[10], according to which the process of excitation of spin waves should become still harder with decrease of temperature. The experimental results can be explained by assuming that the number of intermediary magnons increases because of an increase of their lifetime with decrease of temperature. If the number of intermediaries appreciably exceeds the number of parametric magnons, then the influence of the latter on the reservoir of thermal spin waves, thanks to which the mechanism of hard excitation becomes possible, can be neglected.

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