Two-photon absorption in the microwave band

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The nonlinear susceptibility of two-photon absorption in the microwave band is determined experimentally for the first time. Its value for the investigated yttrium iron garnet samples was found to be $(0.75\pm0.45)\times10^{-5}$ cgs esu. The experiments were carried out at 2400 MHz with the microwave generator operating in the pulsed regime; an open dielectric resonator made of single-crystal strontium titanate was used to increase the sensitivity of the apparatus. The experimental value of the nonlinear susceptibility is half as large as expected from the theoretical estimates, apparently owing to the parametric excitation of spin waves at double the pumping frequency.

Two-photon absorption in the microwave band was first described in[1]. It was impossible there, however, to obtain quantitative data on the susceptibility of this process, owing to its low probability in the microwave band. Two-photon absorption can be relatively easily registered in the optical and radio bands[2,3]. In the former case, lasers make it possible to obtain field intensities comparable with atomic intensities, and in the latter the intensities of the high-frequency magnetic fields should not be small in comparison with the external resonant magnetic field, which is of the order of one Øersted, and it is also easily attainable in practice. In the microwave band the necessary constant magnetic fields reach several thousand Øersted, and this greatly decreases the probability of two-photon absorption. In[1] it was possible to obtain a microwave magnetic field smaller by three orders of magnitude than the external constant magnetic field applied to the ferrite sample. The susceptibility of the two-quantum absorption was approximately six orders of magnitude lower than the linear susceptibility. The measurements in[1] were carried out at an average microwave-source power 15 W, and this led to a strong heating of the sample. In the present study we determined the power and the nonlinear susceptibility of two-photon absorption in spherical samples of single-crystal yttrium-iron garnet (YIG) at 2400 MHz; the measurements were made at room temperature.

THEORY

Two-photon absorption is a process in which a particle absorbs simultaneously two incident photons, each with energy $\hbar\omega$, and is in an excited state separated from the ground state by an energy $2\hbar\omega$[4]. If there is an allowed energy level near $2\hbar\omega$, then the two-photon absorption has a resonant character. As applied to magnetic crystals, this means that the dependence of two-photon absorption on the constant magnetic field H_0 has a resonant character with a maximum near $H_0 = 2\omega/\gamma$, whereas ordinary linear absorption is resonant at $H_0 = \omega/\gamma$. Here γ is the gyromagnetic ratio for the electron spin.

The magnitude of the two-photon absorption can be determined by solving the equation of motion of the magnetization of a ferrite sphere

$$\frac{d\mathbf{M}}{dt}=-\gamma[\mathbf{M}\times\mathbf{H}]+\omega_r\left(\frac{M_0}{H_0}\mathbf{H}-\mathbf{M}\right),\qquad(1)$$

where $\mathbf{M} = \mathbf{z}M_0 + \mathbf{m}$ (M_0 is the saturation magnetization, $\mathbf{m}$ is the alternating magnetization, $m \ll M_0$); $\mathbf{H} = \mathbf{z}H_0 + \mathbf{h}$ ($\mathbf{h}$ is the alternating magnetic microwave field $h \ll H_0$); ω_r is the ferrite relaxation frequency and is equal to $\frac{1}{2}\gamma\Delta H$ (ΔH is the ferromagnetic resonance (FMR) absorption line width, and $\mathbf{z}$ is a unit vector along the z axis).

In the language of classical physics, the two-photon absorption process consists of the following: An alternating field $\mathbf{h}$ of frequency ω produces in the magnetization spectrum, by virtue of the nonlinearity of the motion of $\mathbf{M}$, a second harmonic $\mathbf{m}_2$ of frequency 2ω. The beats between $\mathbf{m}_2$ and $\mathbf{h}$ form a magnetization harmonic that varies at the same frequency as the external field, i.e., ω. Consequently, an additional nonlinear magnetization $\mathbf{m}_1^{\mathrm{n}}$ appears in the magnetization spectrum and varies in synchronism with the external field, absorbing at the same time a power P_2 from the external field:

$$P_2=\left(V\Big/\frac{2\pi}{\omega}\right)\int\limits_0^{2\pi/\omega}\mathbf{m}_1{}^{\mathrm{n}}\frac{d\mathbf{h}}{dt}\,dt,\qquad(2)$$

where V is the volume of the ferrite, and P_2 is the power of the two-quantum absorption of the ferrite.

The way of determining P_2 is clear from the physical picture of the process. It is first necessary to find $\mathbf{m}_2$, by making in (1) the substitution

$$\mathbf{m}=\frac{1}{2}\sum_{n=-\infty}^{\infty}\mathbf{m}_n e^{in\omega t},\qquad \mathbf{h}=\frac{\mathbf{h}_0}{2}e^{i\omega t}+\text{c.c.}$$

and assuming that $m_n \ll m_{n-1}$[5]. Equation (1) is then used to write down an equation for $\mathbf{m}_1$, retaining terms up to third order of smallness inclusive:

$$i\omega\mathbf{m}_1=-\gamma([\mathbf{M}_0\times\mathbf{h}_0]+[\mathbf{m}_1\times\mathbf{H}_0])-\gamma[\mathbf{m}_2\times\mathbf{h}_0^*]+\omega_r\frac{M_0}{H_0}\mathbf{h}-\omega_r\mathbf{m}_1.\qquad(3)$$

The first term in the right-hand side of (3) is due to the usual linear ferromagnetic resonance. The second term is responsible for the two-quantum absorption; it is easily seen that it is precisely this term which ensures the frequency conversion between the external field and the magnetization second harmonic that can be obtained by the method indicated above.

In the resonant case ($H_0 = 2\omega/\gamma$) and in the presence of only two components of the external microwave field h_x and h_z, we obtain

$$|m_{1x}^{\mathrm{n}}|=\frac{\gamma^3|h_{0z}|^2|h_{0x}|M_0}{8\omega^2\omega_r},\qquad |m_{1z}^{\mathrm{n}}|=\frac{\gamma^3|h_{0x}|^2|h_{0z}|M_0}{8\omega^2\omega_r}.\qquad(4)$$

Substituting (4) in (2), we have for the power of the two-photon absorption at the maximum of the absorption line

$$P_2^{max}=\chi''_{\text{res}}\frac{\gamma^2|h_{0x}|^2|h_{0z}|^2V}{4\omega}\equiv\chi_2''\,\omega|h_{0x}|^2|h_{0z}|^2V \qquad (5)$$

where $\chi''_{\text{res}} = M_0/\Delta H$; χ''_2 is the nonlinear susceptibility of the two-photon absorption, and its value in accordance with the definition (5) is

$$\chi_2''=\chi''_{\text{res}}\gamma^2/4\omega^2. \qquad (6)$$

EXPERIMENT

For an experimental investigation of the two-photon absorption, the ferrite sample was placed inside a microwave resonator, and the voltage standing wave ratio (VSWR) of the resonator was then measured at different incident pump powers P and different values H_0. If P_0 or $|H_0 - 2\omega/\gamma| \gg \Delta H$, then the two-photon absorption power was small and the resonator has a VSWR equal to r_0. With increasing power P, the losses in the resonator increase as a result of the two-photon process, the Q of the resonator decreases, and this broadens the resonant curve of the resonator and changes its VSWR, which is now equal to r. It is understandable that a connection exists between the additional losses (in our case equal to P_2) and the relation between r_0 and r. Depending on the coupling coefficient β we have[6]

$$P_2=(r_0/r-1)P_{\text{abs}} \quad \beta\geq 1,$$
$$P_2=(r/r_0-1)P_{\text{abs}} \quad \beta\leq 1, \qquad (7)$$

where P_{abs} is the pump power absorbed in the resonator. For "reflex" resonators, P_{abs} is the difference between the power incident on the resonator P and the power reflected from it. For a flow-through resonator, the relations are more complicated, but for the case of critical coupling ($\beta = 1$), which is convenient in practice, we have $P_{\text{abs}} = P/2$.

It is seen from (5) that P_2 is proportional to the fourth power of the amplitude of the microwave magnetic field in the resonator. Therefore, to observe two-photon absorption we used an open dielectric resonator made of strontium titanate, since it is known[7] that in a dielectric resonator with dielectric constant ϵ ($\epsilon \approx 300$ for strontium titanate) the field amplitudes are larger by approximately $\epsilon^{3/4}$ times than at the same absorbed power in a hollow cavity resonator. In addition, the use of dielectric resonators has one important advantage in that the filling factor of the ferrite in the resonator, which determines the sensitivity of the apparatus, is increased because of the decrease in the geometrical dimensions.

The open dielectric resonator was a disk of 12 mm diameter and 3 mm thickness. A hole in which to place the YIG spheres, of 2.8 mm diameter (the diameter of the investigated ferrite spheres) was drilled along the disk axis. The dielectric resonator with the ferrite was tuned to the pump-generator frequency 2400 MHz. To reduce the temperature rise of the sample, pulsed operation of the microwave generator was used, with pulse duration 2—10 μsec at a repetition frequency 50 Hz.

The experimental setup was the following. The microwave generator was connected through a gate, a variable attenuator, and a double-directional coupler to a symmetrical strip line which served as the excitor for the open dielectric resonator. The coupling between the resonator and the strip line was varied by moving the dielectric resonator inside the strip line. The second end of the strip line was short circuited ("reflex" regime) or connected to a matched load ("transmission" regime). The experiment gave identical results in both cases. The bilateral directional coupler was used to measure the power incident on the resonator and to determine the VSWR of the resonator with the ferrite.

The dielectric resonator with the YIG sample was located inside a permanent magnet producing a constant magnetic field that was double the field value ω/γ at which the FMR takes place. In this field, the linear loss of the ferrite is quite small and practically independent of the constant magnetic field. At the same time, as shown above, the two-quantum absorption near the constant field $H_0 = 2\omega/\gamma$ is maximal, and its rate of change as a function of the change H_0 is also maximal, inasmuch as the two-quantum absorption is described by a Lorentz curve with a center at $H_0 = 2\omega/\gamma$.

The VSWR of the resonator with the ferrite was determined experimentally with a precision attenuator, by comparing the power incident on the resonator with that reflected from it. The ratio r/r_0 needed to calculate the two-photon absorption power with the aid of relation (7) can also be obtained by a simpler method, namely by comparing the reflected signal in the absence of two-photon absorption (this signal is proportional to r_0) and in the presence of this absorption (in which case the signal is proportional to r). In this experimental procedure, the power reflected from the resonator was measured by a detector calibrated against a precision attenuator, was subsequently integrated, amplified with a dc amplifier, and finally registered with an automatic recorder whose sweep was in synchronism with the variation of the constant magnetic field. The incident power was maintained constant. Integration of the signal was necessary because of the pulsed operation of the pump generator. Thus, the reflected power at any point of the resonant curve of the two-photon absorption could be determined from the curves on the recorded chart. Far from the resonant field $H_0 = 2\omega/\gamma$, the automatic recorder shows the value of the reflected power in the absence of two-phonon absorption, so that all the data recorded for the determination of r/r_0 were available.

A typical plot of the VSWR of an open dielectric resonator with a ferrite as a function of the variation of the constant magnetic field is shown in Fig. 1.

From the measured values of r/r_0 it is possible, by using formula (7), to determine the values of the power absorbed by the two-quantum mechanism. Figure 2 shows a plot of the two-quantum absorption power at the maximum of the absorption line against the incident power for a YIG sample of 2.8 mm diameter at $\Delta H = 2$ Oe. It is seen from the figure that the dependence is close to quadratic, in accord with (5).

The results of the experiment show that the employed procedure can yield, at small incident powers (less than 1 kW in pulse) noticeable values of two-quantum absorption (more than 20 W). It should be noted here that the two-quantum absorption is the limiting power generated at nonresonant frequency doubling in ferrites, the efficiency of which reaches 30%[8], i.e., the magnitude of the two-quantum absorption is not less than 30% of the incident power. In this case, however, large ferrite samples are used and the oscillations in the dielectric resonators are strongly perturbed, so that it is difficult to compare the experiments with the theory.

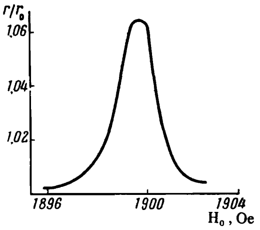

FIG. 1

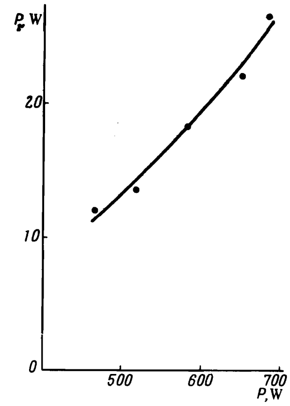

FIG. 2

FIG. 1. Dependence of VSWR of a dielectric resonator with a ferrite on the magnetic field near the value $2\omega/\gamma$; P = 580 W.

FIG. 2. Dependence of the two-quantum absorption power on the incident power. Solid line—quadratic dependence.

Using formula (5) it is possible to calculate from the experimentally determined value of P_2^{max} the nonlinear susceptibility χ_2'' of the two-photon absorption:

$$\chi_2''{}_{\exp} = \frac{P_{2\,\exp}^{max}}{|h_{0x}|^2|h_{0z}|^2V}. \qquad (8)$$

The amplitude of the microwave fields in the resonator are expressed in terms of the power absorbed by the resonator[7], with $|h| = c\sqrt{P_{abs}}$, where c is a constant characterizing the resonator parameters. In our case $c = 1.55$ Oe-W$^{-1/2}$. The value of $\chi_{\exp}''$ determined in this manner turned out to be, in accord with the theory, independent of the incident power and equal to 0.75×10^{-5} cgs esu.

Let us estimate the measurement errors. The accuracy with which the VSWR is measured depends on the accuracy of the precision attenuator, which for our case (we used a DZ-29 polarization attenuator) was better than 1%. The microwave power was measured with an accuracy of 10%. The main contribution to the measurement errors is made by the inaccuracy of the constant c in the formula that connects the field in the resonator with the absorbed power. The theoretical formula for c[7] should be used with great caution, since it does not take into account the presence of the ferrites in the resonator, the presence of an opening in the resonator, etc. We have therefore determined c experimentally from the threshold of the spin-wave instability in ferrites[9]. In this case the measurement accuracy does not exceed 10%. The relative error in the determination of χ_2'', calculated on the basis of the data presented above, is 60%. Thus,

$$\chi_2''{}_{\exp} = (0.75 \pm 0.45)\cdot 10^{-5}\ \text{cgs esu}.$$

At the same time formula (6), which determines the theoretical value of the susceptibility of the two-photon absorption, yields a value approximately twice as large ($\sim 1.9 \times 10^{-5}$ cgsesu). This discrepancy is probably due to the fact that in the derivation of (6) we did not take into account the influence of the spin waves on the two-quantum absorption process[9]. This influence can arrive, for example, as a result of parametric fourth-order processes, in which spin waves are generated with a frequency double the pump frequency[10]. For our case the threshold of this process, according to[10], is on the order of 100 W. This indicates that at P > 100 W the pump energy is consumed not only in two-quantum processes, but also in parametric excitation of spin waves, and this should naturally lead to a decrease of χ_2''.

[1] I. A. Deryugin, V. V. Zaporozhets, and G. A. Melkov, ZhETF Pis. Red. 5, 352 (1967) [JETP Lett. 5, 292 (1967)].
[2] J. L. Hall, D. A. Jennings, and R. McClintock, Phys. Rev. Lett. 11, 364 (1963).
[3] D. N. Klyshko and V. S. Tumanov, Zh. Eksp. Teor. Fiz. 47, 2035 (1964) [Sov. Phys.-JETP 20, 1367 (1965)].
[4] V. M. Faĭn, Fotony i nelineinye sredy (Photons and Nonlinear Media), Sov. radio (1972).
[5] I. A. Deryugin, A. A. Konchits, and G. A. Melkov, Radiotekhnika i élektronika 11, 150 (1966).
[6] E. L. Ginzton, Microwave Measurements, McGraw, 1957.
[7] G. A. Melkov, Radiotekhinika i élektronika 17, 2027 (1972).
[8] G. A. Melkov and A. L. Lutsenko, Radiotekhnika i élektronika 18, 350 (1973).
[9] H. Suhl, J. Phys. Chem. Sol. 1, 209 (1957).
[10] I. A. Deryugin, G. A. Melkov, and V. L. Grankin, Radiotekhnika i élektronika 15, 2583 (1970).

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