

agreement with the values of  $\tau_s$  obtained above for these samples.

It follows from the above results that the dynamic nuclear polarization is most effective in weak magnetic fields if it is due to the dipole-dipole interaction. This is associated with the fact that a reduction in the magnetic field increases the contribution made to the nuclear relaxation process by the transitions involving the rotation of the electron and nuclear spins in the same ( $\Delta m_s = \pm 1$ ,  $\Delta m_I = \pm 1$ ) and opposite ( $\Delta m_s = \pm 1$ ,  $\Delta m_I = \mp 1$ ) directions.

The range of magnetic fields in which nuclei are polarized dynamically as a result of the dipole-dipole interaction of nuclei with electrons trapped by deep levels is a function of the spin relaxation time of these electrons. This makes it possible to use optical pumping in weak magnetic fields in estimating the spin relaxation time of electrons localized at deep levels.

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## Relaxation of longitudinal microwave magnetization in parametric excitation of spin waves in ferrites

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We measured the relaxation frequency of the longitudinal macroscopic alternating magnetization produced in ferrites when spin waves are parametrically excited by parallel pumping. It is observed that this frequency always exceeds the spin-wave relaxation frequency determined from the threshold of the parametric excitation, and depends on many parameters such as the supercriticality, temperature, and sample diameter. To explain the experimental results, it is assumed that phase mismatch contributes to the damping of the longitudinal macroscopic magnetization. This mismatch is a result of the fact that the pump excites a packet of spin waves whose distribution over the eigenfrequencies has a finite half-width. The existing nonlinear theory of parametric excitation of spin waves in ferrites leads only to qualitative agreement with experiment, and great discrepancies appear in the quantitative estimates. The objects of the investigation were single-crystal spheres of yttrium iron garnet at a pump frequency 9370 MHz.

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The most important parameter used in the analysis of parametric excitation of spin waves in ferrites is the relaxation frequency of parametrically excited spin waves (PSW) with wave vector  $k - \gamma_k$ .<sup>1</sup> Since the pump excites a PSW packet that is narrow in terms of  $k$  ( $\Delta k \ll k$ , Ref. 2), parametric spin-wave instability is described in the theory only by one characteristic time  $T_k = 1/\gamma_k$ , which determines, in particular, the threshold value of the microwave magnetic pump field  $h_{thr}$  at which spin-wave instability sets in.<sup>3</sup> However, the experiments described below show that a single time  $T_k$  cannot describe even all the macroscopic characteristics of parametrically regenerated ferrites.

We have measured the characteristic time  $T_{kz} = 1/\gamma_{kz}$

of the free damping of longitudinal microwave magnetization  $m_z(t)$  produced in the case of parallel pumping of spin waves in ferrites:  $m_z(t) = m_z \exp(-2\gamma_{kz}t)$ . The magnetization  $m_z$  is the projection of the vector of the microwave magnetization of the ferrite on the  $z$  axis, along which both the constant and alternating magnetic fields  $H_0$  and  $h$  are directed;  $m_z$  is given by<sup>4</sup>

$$m_z = \sum_k (V_k b_k b_{-k} \exp(2i\omega_k t) + c.c.), \quad (1)$$

where  $b_k$  is the complex amplitude of the spin wave with wave vector  $k$ , and  $\omega_k$  is the PSW frequency and is equal to half the pump frequency  $\omega_p$ . We note that it is precisely the appearance of  $m_z$  that makes possible absorp-

tion of energy by the spin waves from the microwave pump field.

## EXPERIMENT

1. A block diagram of the experimental setup is shown in Fig. 1. A rectangular microwave pulse ( $\omega_p = 2\pi \times 9.37$  GHz) was directed through a number of waveguide elements into a cavity resonator with a ferrite situated in a permanent magnetic field  $H_0$ . The resonator oscillation mode was  $H_{012}$ , and its  $Q$  was  $\sim 10^3$ . The microwave pulse reflected from the resonator was fed through a bidirectional coupler through precision attenuator  $PA_1$  and traveling-wave tube amplifier to a broadband square-law detector and then was registered with a stroboscopic oscilloscope. The required supercriticality  $\zeta = h/h_{thr}$  was set with the aid of a precision attenuator  $PA_2$  (calibration accuracy  $\pm 0.2$  dB); the value of  $h_{thr}$  and the associated<sup>3</sup> PSW relaxation frequency  $\gamma_k = 2\pi g^2 M_0 \omega_p^{-1} h_{thr}$  ( $g$  is the gyromagnetic ratio for the electron spin and  $M_0$  is the saturation magnetization of the ferrite) were determined from the appearance of a "fracture"<sup>3</sup> on the pulse reflected from the resonator, using a power meter accurate to  $\pm 10\%$ . The experiment was performed on spheres of yttrium iron garnet (YIG) in the temperature range 4.2–295 K. The spheres were magnetized along the difficult axis [100], i.e.,  $H_0 \parallel [100]$ . The duration of the pump pulse was 100–400  $\mu$ sec and the repetition frequency was 50 Hz. The trailing edge of the pump pulse at the level 0.1 of its amplitude was  $\sim 30$  nsec, a value ensured by a modulator with a GU-50 high-frequency pentode. A typical waveform of the signal observed on the stroboscopic oscilloscope is shown in Fig. 2.

In the presence of spin-wave instability ( $\zeta > 1$ ) the aforementioned "fracture" becomes observable on the pulse reflected from the resonator at an instant of time  $t_0$  that depends on the sensitivity of the apparatus. This "fracture" is due to the fact that from the instant  $t_0$  to the time  $t_1$  when the pulse is turned off a noticeable fraction of the energy accumulated in the resonator is consumed by excitation of the PSW, as a result of which the loaded  $Q$  of the resonator decreases, and this is accompanied by a change of the coefficient of reflection of the microwave power from the resonator with the ferrite. The form of the signal on the oscilloscope screen after turning off the pump ( $t > t_1$ ) is shown in enlarged scale in Fig. 2(b). The form of this signal is governed by the following circumstances. Under parametric ex-

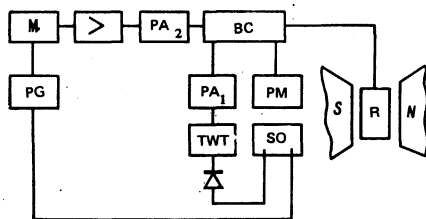


FIG. 1. Block diagram of experimental setup: M—magnetron, PG—generator of synchronizing pulses, PA—precision attenuator, BC—bidirectional coupler, R—resonator with ferrite in a cryostat and in a magnetic field, TWT—traveling-wave-tube amplifier, PM—power meter, SO—stroboscopic oscilloscope.

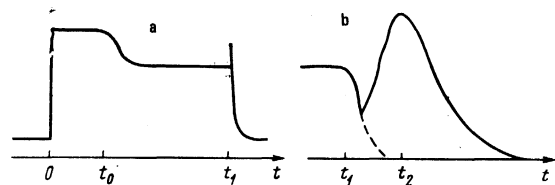


FIG. 2. a) Waveform of pulse reflected from the resonator with the ferrite, b) trailing edge of pulse reflected from the resonator with the ferrite.  $t_0$ —instant of appearance of the "fracture" on the reflected pulse;  $t_1$ —instant when the pump pulse is turned off;  $t_2$ —end of transient process after turning off the pump. The dashed line shows the transient in the absence of parametric excitation of the spin waves.

citation of spin waves, a field  $h$  equal to the sum of the unperturbed field  $h_0$  and the sample emission field  $h_m$  due to the magnetization  $m_x(t)$ , is present in the cavity resonator with the ferrite:  $h = h_0 + h_m$ .<sup>4,5</sup> The amplitude of the signal reflected from the resonator is proportional to the amplitude of the self-consistent field  $h$ ; since a square-law detector is used, the signal on the oscilloscope screen (and consequently on Fig. 2) is proportional to  $h^2$ . In parallel pumping, the phases of all three fields  $h$ ,  $h_m$ , and  $h_0$  are different and can be determined with the aid of the nonlinear theory of parametric excitation of spin waves in ferrites.<sup>1</sup>

A typical time vector diagram of the fields in the resonator ( $h$ ,  $h_m$ , and  $h_0$  are parallel to one another and to  $H$  in space) is shown in Fig. 3.<sup>5</sup> When the pump is turned off, the fields  $h_0$  and  $h_m$  attenuate, and with them the resultant self-consistent field  $h$  also attenuates and changes its phase. If  $h_0$  attenuates much more rapidly than  $h_m$ , and it will be seen from the following that this is the situation in the experiment, then, according to Fig. 3, the phase of the field  $h$  will rotate towards the phase of  $h_m$  after the pump is turned off, while the amplitude of  $h$  will first decrease and then increase and the equality  $h = h_m$  will be reached. The same behavior will be exhibited in time also by the microwave signal reflected from the resonator, as is in fact shown in Fig. 2(b). The rate of damping of the unperturbed field  $h_0$  is determined by the time constant of the cavity resonator and by the slope of the trailing edge of the pulse.

The picture of the damping of  $h_0^2$  with time, shown dashed in Fig. 2, can be observed on the oscilloscope screen in the absence of parametric excitation of spin

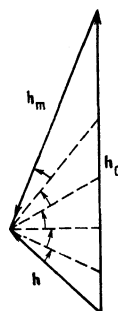


FIG. 3. Explanation of the waveform of the pulse in Fig. 2b. Vector diagram of microwave magnetic fields acting in the resonator on the ferrite. The dashed lines show the changes of the self-consistent field  $h$  due to the damping of  $h_0$  at  $h_m = \text{const}$ .

waves, i.e., at  $h < h_{thr}$ . It is seen from Fig. 2(b) that after the pump is turned off at the instant of time  $t_1$  the transient in the resonator due to the damping of  $h$  is significant only up to the instant  $t \lesssim t_2$ ; in our experimental setup  $t_2 - t_1 \lesssim 80$  nsec.

At  $t > t_2$ , a signal due to the exponentially damped alternating longitudinal magnetization  $m_z(t)$  is observed on the oscilloscope screen. The damping constant of this signal on the oscilloscope depends on the characteristics of the detector, and in the case of an ideal square-law detector it is equal to  $4\gamma_{kz}$ , so that  $\gamma_{kz}$  can be determined from the measurements by analyzing log-log curves similar to those shown in Fig. 2(b). Since, however, no real detector is ideal, this method leads to large errors in  $\gamma_{kz}$ . We used therefore a different method based on the use of a precision attenuator  $PA_1$  graduated in decibels with accuracy  $\pm 0.2$  dB. In this method the characteristic of the detector is of no importance. The gist of this method is that all the measurements are performed at a constant microwave power incident on the detector. To this end, an arbitrary constant reference level is chosen on the oscilloscope screen, where a picture similar to that of Fig. 2(b) is observed. By varying the attenuation  $L$  (in dB) of the attenuator  $PA_1$ , various points due to the signal reflected from the resonator and attenuating exponentially with time are made to coincide with this level at  $t > t_2$ .

The dependence of the attenuation  $L$  at which any particular point of the exponentially damped signal is aligned with a chosen constant level on the time  $t$  corresponding to this point is in fact that  $m_z(t)$  plot in logarithmic coordinates. An example of such a plot is shown in Fig. 4. It is seen that at  $t \lesssim t_2 + 0.2$  sec the plot is a straight line, meaning that  $m_z(t)$  is damped in time exponentially with an exponent  $2\gamma_{kz}$  determined by the slope,  $\gamma_{kz} \cdot 40 \log e$ , of the straight line. It is seen from Fig. 4 that for a time  $t \lesssim t_2 + 0.2$  sec the plot of  $L$  against  $t$  deviates from a straight line, meaning that the  $m_z(t)$  damping is not exponential. This may be due to the influence of the transients in the resonator as well as to nonlinear interactions between the spin waves; these interactions are significant at large PSW amplitudes and can lead, in particular, to changes of the dynamic susceptibility of the ferrite and to returning of the resonator. Therefore all the measurements of  $\gamma_{kz}$  should be carried out only on the linear section of the plot of  $L$

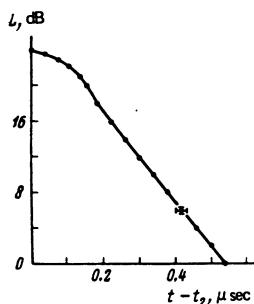


FIG. 4. Dependence of the damped longitudinal magnetization of the ferrite on the time (see Fig. 2b at  $t > t_2$ ) in logarithmic coordinates. Sample—YIG sphere of diameter 2.5 mm,  $\zeta = 1.58$ , and  $\gamma_k = 1.15$  MHz.

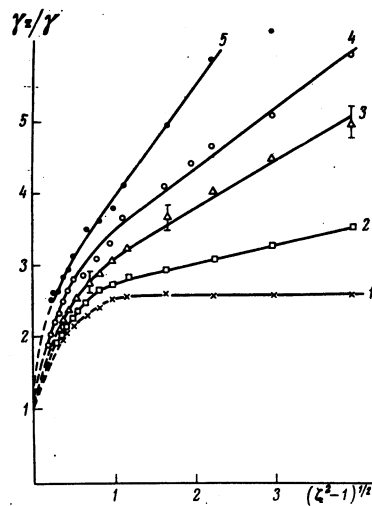


FIG. 5. Dependence of the relaxation frequency of the longitudinal magnetization on the pump level. Sample—YIG sphere of 2.5 mm diameter. Curves: 1— $T = 295$  K,  $\gamma_k = 1.15$  MHz; 2— $T = 20$  K,  $\gamma_k = 0.6$  MHz; 3— $T = 12$  K,  $\gamma_k = 0.37$  MHz; 4— $T = 5.5$  K,  $\gamma_k = 0.2$  MHz; 5— $T = 4.2$  K,  $\gamma_k = 0.14$  MHz. PSW wave vector  $k \approx 10^5$  cm $^{-1}$ ,  $H_0 \parallel [100]$ .

against  $t$ .

2. Experimental plots of the relative relaxation frequency of the longitudinal microwave magnetization against the parameter  $(\zeta^2 - 1)^{1/2}$  that characterizes the pump level are shown in Fig. 5 for one of the investigated YIG spheres. An analysis of the curves in Fig. 5 makes it possible to establish certain general regularities. We note first that  $\gamma_{kz} > \gamma_k$  always, i.e., the relaxation of  $m_z$  is faster than the relaxation of the number of the PSW. Furthermore  $\gamma_{kz}$ , unlike  $\gamma_k$ , is not a constant of the material but depends on the supercriticality and on the temperature. It can furthermore be noted that the ratio  $\gamma_{kz}/\gamma_k$  consists of two terms. One depends little on the temperature and increases sharply at small supercriticalities ( $(\zeta^2 - 1)^{1/2} < 1$ ,  $\zeta^2 < 2$ ), and the other increases in proportion to  $(\zeta^2 - 1)^{1/2}$  with a proportionality coefficient that depends significantly on the temperature  $T$ :

$$\gamma_{kz}/\gamma_k = a + \alpha(\zeta^2 - 1)^{1/2}. \quad (2)$$

In (2) the coefficients  $a = a(\zeta)$  and  $\alpha = \alpha(T)$ .

To determine the nature of the coefficients  $a$  and  $\alpha$ , empirical relations similar to (2) were obtained for single-crystal YIG of various diameters (from 1.06 to 3.5 mm) at different values of the constant magnetic field  $H_0$ , corresponding to excitation of PSW with a wave vector  $10^5$  cm $^{-1} < k < 3 \times 10^5$  cm $^{-1}$ . It was established that the value of  $\alpha$  is determined by size effects that depend on the ratio of the mean free path  $\lambda$  of the parametrically excited spin waves to the radius  $r$  of the investigated sphere. The  $\alpha(T)$  plot shown in Fig. 5 is determined by the growth of the mean free path with decreasing temperature, owing to the increased lifetime  $T_k = 1/\gamma_k$  of the spin waves. The experimental  $\alpha(\lambda/r)$  plot is shown in Fig. 6(a); it is seen that at  $\lambda/r \leq 0.4$  we have  $\alpha \approx 6.5\lambda/2r$ , and with further increase of  $\lambda$  the  $\alpha(\lambda/r)$  curve flattens to a value  $-1.4$ . The coefficient  $a$  as a function of the pump parameter  $(\zeta^2 - 1)^{1/2}$

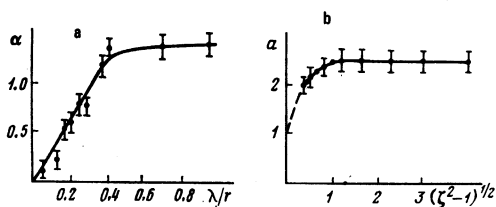


FIG. 6. a) Dependence of the coefficient  $\alpha$  in (2) for different single-crystal YIG spheres on the ratio of the mean free path  $\lambda$  of the PSW to the sample radius  $r$ ; b) dependence of the coefficient  $a$  in (2) on the pump for a YIG sample of 2.5 mm diameter.

is shown in Fig. 6(b). At  $\zeta^2 < 2$  the value of  $a$  increases sharply to  $\sim 2.5$ , where it remains approximately constant at large supercriticalities. The coefficient  $a$  depends little on size effects and changes by several dozen percent over the entire range of variation  $\lambda/2r$  ( $a$  decreases with increasing  $\lambda/2r$ ).

## DISCUSSION OF RESULTS

1. To explain the experimentally observed character of the damping of the longitudinal high-frequency magnetization  $m_z(t)$  we can advance the following arguments. It is seen from (1) that  $m_z$  is determined not only by the number  $\sum_k b_k + b_k$  of the PSW, but also by their phase shifts, so that the damping of  $m_z$  after turning off the pump is determined not only by the damping of the number of the PSW, but also by randomization of the phase shifts  $\psi_k$  of the spin waves; this randomization was the same for all PSW during the time of action of the pump, i.e., in the general case  $\gamma_{kz} > \gamma_k$ , as we have seen above to be indeed the case in experiment. The cause of the randomization of the PSW phase after the pump is turned off may be that parametric pumping excites spin waves whose natural frequencies  $\omega_k$  are not strictly equal to half the pump frequency, but are distributed in a certain finite interval about  $\omega_p/2$ . If we assume a Lorentz distribution with half-width  $\Delta\omega_k$ , then, changing to integration, we can obtain with the aid of (1)

$$m_z(t) = \frac{2}{\pi} m_z(0) \exp(-2\gamma_k t) \int_0^\infty \frac{\Delta\omega_k \cos(2\omega_k t + \psi_k) d\omega_k}{(\omega_k - \omega_p/2)^2 + \Delta\omega_k^2} \approx m_z(0) \exp[-2(\gamma_k + \Delta\omega_k) t] \cos(2\omega_p t + \psi_k). \quad (3)$$

From (3) we have for  $\gamma_{kz}/\gamma_k$ :

$$\gamma_{kz}/\gamma_k = 1 + \Delta\omega_k/\gamma_k. \quad (4)$$

Comparing (4) with (2) we obtain a relation for the experimental determination of  $\Delta\omega_k$ :

$$\Delta\omega_k/\gamma_k = a - 1 + \alpha(\zeta^2 - 1)^{1/2}. \quad (5)$$

2. We now analyze the half-widths  $\Delta\omega_k$  of the PSW distribution in the natural frequencies, obtained with the aid of (5), for the case when the size effects are small ( $\alpha \rightarrow 0$ ), i.e., for the case of an infinite ferrite sample, which is usually considered in the theory.<sup>1</sup> According to (5) and Fig. 6(b) we have  $\Delta\omega_k \approx 1.5\gamma_k$  at  $\zeta^2 \geq 2$ . Such a strong frequency broadening of a packet of PSW greatly contradicts the theoretical relation

$$\Delta\omega_k = \gamma_k \left[ \frac{\gamma_k}{k \partial \omega_k / \partial k} (\zeta^2 - 1) \right]^{1/2}, \quad (6)$$

obtained from the nonlinear theory.<sup>1</sup>

According to (6) the value of  $\Delta\omega_k$  at  $\zeta^2 \approx 2$  is only  $0.1\gamma_k$ . Moreover, even the threshold of the excitation of the spin waves with  $\omega_k = \frac{1}{2}\omega_p \pm 1.5\gamma_k$  is reached in the linear theory at a supercriticality  $\zeta^2 = 6.25$ .<sup>3</sup> We can propose two explanations for the rapid broadening of the PSW packet relative to the natural frequencies: (1) The finite dimensions of the sample make it possible for a pair of spin waves with  $k_1 \neq k_2$ ,  $\omega_{k_1} \neq \omega_{k_2}$  to be excited parametrically, even though as before we have  $\omega_{k_1} + \omega_{k_2} = \omega_p$ . This process has been well investigated for magnetostatic waves,<sup>3</sup> where excitation of waves with frequencies that differ from  $\omega_p/2$  by  $\sim \gamma_k$  is perfectly possible following a small increase of the threshold.

(2) Immediately beyond the instability threshold, inhomogeneous collective oscillations are produced in the PSW system and lead to an increase of  $\Delta\omega_k$ .<sup>1</sup> The amplitude of these oscillations increases rapidly with increasing supercriticality to  $\zeta^2 \approx 2$ , and then saturates. Evidence favoring this explanation is provided by experiments performed on the YIG spheres magnetized along the easy axis [111]. In this case the intensity of the self-oscillations is maximal, as is also the value of  $\Delta\omega_k$ . At  $\zeta^2 = 4$  we have  $\Delta\omega_k \approx 6\gamma_k$ . This explains in addition the already mentioned fact that the coefficient  $a$  decreases with increasing  $\lambda/2r$ , since it is known that size effects decrease the amplitude of the self-oscillations of the magnetization.<sup>6</sup> The role of effects connected with the collapse of the spin waves and with other related phenomena due to the large spin-wave amplitude in the absence of a pump<sup>1</sup> is apparently small, since such processes evolve within a time  $\sim T_k$ , and this would lead to a deviation of  $m_z(t)$  from the exponential law, something not observed in experiment.

3. The dependence of the parameter  $\Delta\omega_k$  of the PSW distribution with respect to the natural frequencies in (5) on the size effects is due apparently to two-magnon scattering of the spin waves by inhomogeneities of the crystal, principally by surface inhomogeneities.<sup>6-8</sup> This is confirmed by the experimentally observed dependence of the coefficient  $\alpha$  on the surface finish of the ferrite, namely,  $\alpha$  increases with increasing grain dimension of the abrasive paste used to polish the samples. The existing theory<sup>7</sup> predicts a dependence of  $\Delta\omega_k$  on the two-magnon scattering; this dependence can be characterized by the parameter  $\gamma_{ks}$  of the scattering of the PSW by the inhomogeneities. From Ref. 7 we can obtain

$$\frac{\Delta\omega_k}{\gamma_k} = \left[ (\zeta^2 - 1)^2 \frac{\gamma_k \gamma_{ks}}{(k \partial \omega_k / \partial k)^2} \right]^{1/2}. \quad (7)$$

In the case of strong size effects ( $T < 12$  K) the experimental data on Fig. 5 can indeed be approximated in accordance with (7), within the limits of experimental accuracy, by the relation  $\Delta\omega_k/\gamma_k \sim (\zeta^2 - 1)^{1/4}$ , but a quantitative agreement with (7) calls for unjustifiably large values  $\gamma_{ks} > 10^3\gamma_k$ , strongly contradicting other experimental results,<sup>7,8</sup> that yield  $\gamma_{ks} \approx \gamma_k$  for the considered experimental conditions.

4. The foregoing results indicate that the model based on randomization of the PSW phases, proposed to explain the relaxation of  $m_z$ , encounters certain difficul-

ties. Moreover, the model itself contains one unclear point. It is quite doubtful, for example, that in all the cases a Lorentz distribution of the PSW in the natural frequencies is realized [only then does (3) lead to an exponential damping of  $m_x(t)$ ], whereas in experiment one always observes an exponential behavior after the termination of the transient. It is therefore possible that the observed effects are due not to randomization of the PSW phases, but to entirely different mechanisms, which could not be established in the present study. We note only that these mechanisms cannot be connected with radiative damping due to the reaction of the field in the resonator on  $m_x$  and due to the inhomogeneity of the field  $H_0$ , inasmuch as these mechanisms were studied in detail and subsequently excluded by choosing small samples and using homogeneous constant magnetic fields. On the other hand, if the  $m_x(t)$  relaxation model based on the phase randomization is valid, this means that the presently existing nonlinear theory of parametric excitation of spin waves,<sup>1,7</sup> must be improved, especially in the presence of self-oscillations of the magnetization and of two-magnon scattering of the PSW by inhomogeneities of the sample. Only this will make it possible, by using the procedure proposed here to determine the frequency of the relaxation of the longitudinal high-frequency magnetization, to determine experimentally such important ferrite characteristics as the parameter  $\gamma_{ks}$  of spin-wave damping by the in-

homogeneities, the spectrum and amplitude of the inhomogeneous oscillations of the magnetization, and even the value  $\gamma_k$  of the linear damping of the PSW, which is very difficult to determine from the threshold of the instability of imperfect crystals.<sup>7</sup>

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## Experimental investigation of the nature of photoelectric phenomena in KDP and DKDP crystals

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A specially developed procedure was used to determine the character of the photoresponse in the dielectric crystals KDP and DKDP to radiation in the transparency band. It is shown that, depending on the experimental conditions on the composition of the crystal, the photoresponse is due mainly to effects of impurity photoconductivity on the nonstationary heating of the crystal lattice, to generation of density oscillations, and to nonlinear optical rectification. It is demonstrated that from the measured photoresponse parameters it is possible to obtain new information on the characteristics of the crystal. In particular, the value of  $(\partial\epsilon/\partial T)$ , of the KDP crystal was measured at low frequencies.

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### 1. INTRODUCTION

Dielectric crystals are widely used in nonlinear-optics systems and instruments. However, the study of the structure-sensitive properties, which are responsible for many aspects of the behavior of these materials in laser fields, has not been properly developed as yet. Thus, for most "nonlinear" crystals there is practically no information on the dynamics of the electrons in the allowed bands, on the properties of the defect and impurity states, and others.<sup>1</sup> This is due to the specific

nature of dielectrics, which hinders direct application of procedures developed and verified for the corresponding purposes in semiconductors. However, in analogy with semiconductors, one should expect an investigation of photoelectric phenomena that accompany the interaction between laser radiation and dielectric crystals to be able to fill the existing gaps. Of course, the photoresponse of wide-band crystals to radiation with energy quantum  $\hbar\omega$  much less than the band gap<sup>2</sup>  $E_g$  does in fact have a number of specific features connected with the low dark conductivity, low photocarrier concentration,