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STATISTICAL EFFECTS CONNECTED WITH THE GENERATION OF OPTICAL HARMONICS

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Results of an experimental and theoretical investigation of statistical effects appearing during generation of the second harmonic in optically transparent crystals are reported. It is established experimentally that under real conditions the correlation coefficient between the second-harmonic power P_2 and the square of the power P_1 of the fundamental radiation emitted by a solid state laser differs from unity, and the proportionality factor K in the equation $P_2 = KP_1^2$ is a random quantity. In order to explain these effects in the prescribed-field approximation of the fundamental radiation, a theory of generation of optical harmonics in the field of randomly modulated waves is developed, which takes into account spatial as well as temporal incoherence of the fundamental radiation. The spatial dimensions characterizing the generation of optical harmonics by a bounded, randomly-modulated beam in an anisotropic medium are determined. It is found that when the optical harmonics are generated by means of a ruby or neodymium-glass laser the main sources of excess fluctuations of the second harmonic power are fluctuations of mode phases, mode number, and angular divergence of the fundamental radiation. Experiments on generation of optical harmonics and mixing of frequencies with the aid of non-laser light sources are briefly discussed. It is noted that space-incoherence effects are important in this case.

1. INTRODUCTION

1. Preliminary Remarks

The rapid development of nonlinear optics has made timely an investigation of problems involving the interaction between randomly modulated electromagnetic waves in nonlinear dispersive media. Even relatively early experiments on the generation of optical harmonics, carried out with the aid of pulsed solid-state lasers, disclosed sufficiently "coarse" effects of statistical nature. We are referring here to the so-called "excess" fluctuations of second-harmonic power generated in crystals by ruby or neodymium-glass lasers (see [1, 2]). The statistical reduction of the experimental data has shown that the fluctuations of the second-harmonic power P_2 cannot be attributed solely to fluctuations of the power of the fundamental radiation P_1 (in solid-state lasers operating at room temperature, the latter, as is well known, are large).

Problems involving fluctuation phenomena in the case of nonlinear transformations of electromagnetic oscillations have already been considered in detail for nonlinear elements with lumped parameters (see, for example, [3]). However, the wave character of the nonlinear optical effect leads to the appearance of many singularities which strongly distinguish them from nonlinear effects in systems with lumped parameters. Therefore in the case of nonlinear transformations of optical radiation the analysis of fluctuation phenomena must be in many respects carried out anew.

In the papers cited above,[1,2] the excess fluctuations of the second-harmonic power were interpreted on the basis of the simplest one-dimensional theory in a quasi-static approximation. It must be noted that the experimental data cited in these papers are very skimpy. We therefore consider it advantageous to present a more detailed analysis of the sources of fluctuations of harmonic power, taking into account the effects of temporal and spatial incoherence of the fundamental radiation and the limited dimensions of the beam of fundamental radiation, to disclose the relative role of the local and accumulating effect, etc. The latter is especially desirable because interest in nonlinear optical effects in the field of a non-laser source has increased recently. We present below the results of an investigation aimed at assessing the influence of the statistics of the fundamental radiation on the process of generation of optical harmonics. For cases when the harmonics are generated with the aid of a laser, the results of the theory are compared with the authors' experiments. We discuss the difficulty of obtaining accumulating nonlinear effects in experiments with non-laser sources.

2. Statistics of Fundamental Radiation

A theoretical investigation of the statistical effects occurring in the generation of harmonics should be based, obviously, on a certain statistical model of the fundamental radiation. If we are dealing with the generation of harmonics by lasers, the results of investigations carried out to date can be summarized, with accuracy adequate for our problem, in the following manner.[1)] The radiation from a pulsed solid-state laser constitutes a spherical wave (in a laser with flat mirrors, modes are excited which correspond to a spherical resonator, this being connected with the influence of the inhomogeneities—see [5-7]). The temporal spectrum of this wave consists of a set of practically equidistant modes [8-11] the transverse structure of which in a given pulse is practically the same (see [5,9]). Upon excitation of higher transverse modes in a spherical resonator, the directional distribution of the laser radiation is in the form of a spot of almost uniform intensity (its fine structure constitutes a set of many spots). The number of spots q, which depends on the type of transverse oscillation excited, does determine essentially the angular divergence 2ϑ of the laser beam. The value of 2ϑ measured at an intensity level of 0.03 of the maximum, is given by an expression of the type (see [7])

$$2\vartheta = 2\lambda(2q+1)/\pi D, \tag{1}$$

where λ is the wavelength of the laser radiation, D is the dimension of the excitation region, and q is the transverse index.

In accordance with the foregoing, the radiation field of a laser in a certain direction, defined by the angles α and β (α is measured from the longitudinal axis of the resonator, and β is measured in a plane perpendicular to the resonator axis), can be written in the form

$$\mathbf{E}_1(\alpha, \beta, \mathbf{r}, t) = \mathbf{e}_1 \sum_{m=1}^{N} A_{1m}(\alpha, \beta, \mathbf{r}, t) \times \cos[\omega_{1m}t - \mathbf{k}_{1m}(\alpha, \beta)\mathbf{r} + \varphi_{1m}(t)]. \tag{2}$$

Here and throughout the index 1 denotes that the corresponding quantity pertains to the fundamental-radiation field; A_{1m}, $\mathbf{k}_{1m}$, and φ_{1m} are the amplitude, wave vector, and phase of the m-th longitudinal mode; $\omega_{1m} = \omega_0 + (m-1)\Delta\omega$ is the frequency of the m-th mode, and N is the number of modes. In the general case the quantities A_{1m} depend on the modulus of the radius vector l, $A_{1m} \sim l^{-1}$. However, if we are interested in the radiation field at intervals of l that are small compared with the distance from the phase center of the radiation L (L, L + l; $l/L \ll 1$), we can neglect the foregoing dependence.

In (2), the phases $\varphi_{1m}(t)$ and the amplitudes $A_{1m}(t)$ are random functions of the time. It will be convenient to represent the phase of an arbitrary mode $\varphi_{1m}(t)$ in the form of two terms

$$\varphi_{1m}(t) = \varphi_{1m}^{(nc)}(t) + \varphi_{1m}^{(c)}(t). \tag{3a}$$

Here $\varphi_{1m}^{(nc)}$ are the noncorrelated phase components, connected with the direct action of the noise on the laser; $\varphi_{1m}^{(c)}$ are the correlated components of the phases, connected with the technical irregularities in the laser resonator, in the active me-

[1)]Data on statistical properties of radiation of ordinary non-laser light sources can be obtained in[4].

dium, etc., and also with the non-isochronous behavior (generally speaking, the time variation of the phase of the m-th mode depends to a certain degree on the amplitudes of all the modes, see [12]). By virtue of the foregoing, the N-dimensional distribution function of the phases $\varphi_{1m}^{(nc)}$ is equal to

$$w_N(\varphi_{11}^{(nc)}, \varphi_{12}^{(nc)}, \ldots \varphi_{1N}^{(nc)}) = w_1(\varphi_{11}^{(nc)}) \ldots w_1(\varphi_{1N}^{(nc)});$$

$$w_1(\varphi_{1m}^{(nc)}) = 1/2\pi. \tag{3b}$$

Inasmuch as we are interested primarily in the excess power fluctuations $P_2(t)$, which are not connected directly with the fluctuations of P_1, we assume for simplicity that the amplitudes of the fundamental radiation are fully correlated:

$$A_{11}(t) = A_{12}(t) = \ldots = A_{1N}(t) = A_{10}(t). \tag{4}$$

Assuming that transverse modes of sufficiently high orders are excited, we assume that the angular distribution of the amplitude is normal and is the same for all axial modes:

$$A_{1m}^2(\alpha, \beta, t) = A_{10}^2(0, t) \exp\{-\alpha^2/2\theta_0^2\}. \tag{5}$$

Here θ_0 is the angular divergence of the radiation, measured at an intensity level 0.61 of maximum ($\theta^2 = \vartheta^2/7$ (see (1)), and the dependence on the angle drops out by virtue of the proposed spherical symmetry of the radiation.

At the present time, the two most frequently used pulsed solid-state laser regimes are the so-called free-generation regime, in which the laser radiation consists of a random sequence of "spikes" of durations τ_p ranging approximately from 0.1 to 1 μsec, and the Q-switched regime, in which single pulses with $\tau_p \approx 10$ nsec are produced. In both regimes, the number of produced axial modes N is in the general case a random variable from pulse to pulse (the corresponding data are given for the spike regime in [9-11, 5], and for the Q-switched regime in [13]). The statistics of the number of excited modes will be characterized henceforth by a distribution function $\mathcal{P}(N)$:

$$\sum_{N=1}^{N_{max}} \mathcal{P}(N) = 1,$$

where N_{max} is the maximum number of modes excited in a given laser.

According to [5, 7], the number of the transverse mode also fluctuates in general from spike to spike. Analogous fluctuations are observed also in the Q-switching regime. In our model we shall take account of these fluctuations by introducing the distribution density w for the divergence θ_0 in such a way that

$$\int_0 w(\theta_0)\, d\theta_0 = 1.$$

Finally, the amplitude distribution function $w(A_{10})$ for the spike regime (especially for a laser operating at room temperature) is most naturally assumed to be of the Rayleigh type:[2]

$$w(A_{10}) = A_{10} A_0^{-2} \exp\{-A_{10}^2/2A_0^2\}. \tag{6}$$

3. MEASURABLE STATISTICAL CHARACTERISTICS

The second-harmonic radiation power of a laser is usually recorded with the aid of systems, for example, photomultipliers, which have time constants τ_r which greatly exceed $\Delta\omega^{-1}$. The values of the corresponding photomultiplier currents will be

$$I_1 = a_{10} P_1 = a_{10} \iint \langle E_1^2(\alpha, \beta, t)\rangle \sin\alpha\, d\alpha\, d\beta, \tag{7a}$$

$$I_2 = a_{20} P_2 = a_{20} \iint \langle E_2^2(\alpha, \beta, t)\rangle \sin\alpha\, d\alpha\, d\beta. \tag{7b}$$

Here a_{10} and a_{20} are the apparatus constants for the fundamental-radiation and second-harmonic channels; the integration is in the plane of the photocathodes and the angle brackets denote averaging over the time τ_r.

Inasmuch as the ruby-laser beam divergence 2ϑ does not exceed 20–30′,[9, 14, 15] and for a neodymium glass laser it is even smaller (~ 10′), we can replace sin α in (7) by α. Substituting (2) in (7) and taking (4) and (5) into account, we obtain for the current I_1

$$I_1 = a_1 F_1(\theta_0) V_1(A_{10}(0, t)),$$

$$V_1 = \sum_{m=1}^{N} A_{1m}^2(0, t) = N A_{10}^2(0, t), \tag{8}$$

where

$$F_1(\theta_0) = \int_0^{2\pi} d\beta \int_0^{\infty} \exp(-\alpha^2/2\theta_0^2)\, \alpha\, d\alpha = 2\pi\theta_0^2; \quad a_1 = a_{10}/2.$$

In the region of small transformation coefficients ($P_2 \ll P_1$), in accordance with dynamic theory of second-harmonic generation, we should have

$$I_2 = K I_1^2, \tag{9}$$

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[2]It is appropriate to note here that in a laser operating in the continuous mode the amplitude distribution function is, of course, gaussian. Strictly speaking, a gaussian distribution with a time-dependent mean value obtains also for the Q-switching mode; here however, we shall use expression (6) throughout for the sake of simplicity.

where K is a constant. Actually, as already indicated, the quantity K is a random one and consequently the fluctuations of I_2 are determined not only by the level of the fluctuations of I_1^2, but also by other factors ("excess" fluctuations). The total dispersion $\overline{\Delta I_2^2}$ of the quantity I_2, can thus be represented in the form of two terms $\{\overline{\Delta I_2^2}\}^{(c)}$—a term which is fully correlated with I_1^2, and $\{\overline{\Delta I_2^2}\}^{(nc)}$—a term which is not correlated with I_1^2.

The quantity characterizing most completely the level of the uncorrelated ("excess") fluctuations is, as follows from mathematical statistics (see, for example, [16]), the coefficient of correlation between I_2 and I_1^2:

$$R = \{\overline{I_2 I_1^2} - \overline{I_2}\,\overline{I_1^2}\} / \{[\overline{I_2^2} - (\overline{I_2})^2]^{1/2} [\overline{I_1^4} - (\overline{I_1^2})^2]^{1/2}\}. \quad (10)$$

Consequently we shall pay considerable attention in this paper to an investigation of R. At the same time, especially when using experimental data on the generation of harmonics for the measurement of nonlinear parameters of a medium, interest attaches also to an investigation of the statistics of the proportionality coefficient K in (9). (The statistics of this very quantity was investigated in [1,2]). The level of the fluctuations of K is

$$\xi = \{\overline{K^2} - (\overline{K})^2\}^{1/2} (\overline{K})^{-1}. \quad (11)$$

2. EXPERIMENT. STATISTICAL REDUCTION METHOD

Our experiments were made with a laser operating in the free-generation mode (a diagram of the setup and its description are contained in [1]). The ruby laser radiation was guided through a KDP (KH_2PO_4) crystal oriented in the direction of synchronism relative to the incident radiation. The spikes of the second-harmonic laser radiation, obtained on the screen of a two-beam oscilloscope, were photographed on sensitive film. The gains of the fundamental and second-harmonic channels were chosen such that the amplitudes of the spikes did not go beyond the linearity range of the gains of the photomultipliers and of the oscilloscope amplifiers.

An oscillogram of the laser second-harmonic spikes is shown in Fig. 1. A sample set of fundamental and harmonic spikes was automatically obtained as the oscilloscope was triggered by one of the laser spikes. The number of spikes in the sample was determined by the duration of the oscilloscope sweep and by the ability to resolve individual spikes; in our experiment the number of spikes in the sample was approximately 40.

For a given sample, we determined the correlation coefficient and the proportionality coefficient K. The sample oscillograms were reduced in the following manner. Using an arbitrary scale, we measured the powers of the second-harmonic spikes I_{2i} and of the fundamental radiation I_{1i} (i—number of spike). The coefficient K as given by the data of the i-th measurement is

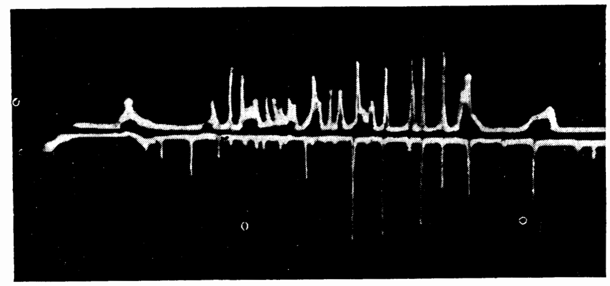

FIG. 1. Oscillogram of ruby-laser emission spikes (top) and of the second harmonic in a crystal (KDP) (bottom), obtained with the aid of a double-beam oscilloscope (the harmonic spikes are narrower than the fundamental ones).

$$K_i = I_{2i} / I_{1i}^2 \quad (12)$$

and is a random quantity from spike to spike. The calculation of the mean value of K, and also of the mean values of I_2, $\overline{I_1^2}$, and the correlation $\overline{I_2 I_1^2}$ in (10) was carried out for the entire volume of the sample; for example,

$$\overline{I_1^2} = \sum_i I_{1i}^2 / M$$

(here M is the number of spikes in the sample). For each calculated value of R we determined the confidence interval ΔR characterizing with a probability of 0.997 the difference between the sample and the true values of R. The probability of the deviations of the correlation coefficient was estimated with the aid of the κ criterion (see, for example, [17]). The experimental values of the correlation coefficient R with the confidence intervals and of the values of ξ (11) are listed in Table I.

3. THEORY OF GENERATION OF SECOND HARMONIC IN THE FIELD OF A MODULATED WAVE. GENERAL ANALYSIS

The laser radiation, as follows from the analysis in Secs. 1 and 2, has a complicated structure: besides the temporal non-monochromaticity (due to the finite duration of the fundamental-radiation pulse and the presence of several longitudinal modes), the laser radiation, which constitutes a beam of finite divergence, has also spatial non-monochromaticity. Strictly speaking, the spatial

Table I. Experimental values of the correlation coefficient R and of the relative fluctuations ξ of the coefficient K.

№	$R \pm \Delta R$	ξ	№	$R \pm \Delta R$	ξ
1	1.00±0.00	0,19	18	0,78±0,20	0,29
2	0.34±0.08	0.31	19	0,99±0,01	0,15
3	1.00±0.00	0.17	20	0,61±0,30	0.31
4	0,94±0.08	0.30	21	0,74±0,24	0,22
5	0.88±0.11	0,24	22	0,66±0,32	0.50
6	0,99±0,05	0,29	23	0,74±0,25	0.38
7	0,96±0,04	0,20	24	0,66±0.30	0,39
8	0,90±0.01	0,30	25	0,64±0.38	0.40
9	0,84±0.15	0,23	26	0,86±0,16	0,18
10	0,95±0,05	0,26	27	0,99±0,01	0,10
11	1,00±0,00	0,28	28	0,70±0,27	0.23
12	0,62±0.30	0.38	29	0,70±0,34	0,20
13	0.55±0,31	0,47	30	0.55±0.42	0,47
14	0,68±0,28	0,20	31	0.37±0.00	0,16
15	0,60±0,35	0.25	32	0,96±0,01	0,18
16	0,81±0.08	0,10	33	0,85±0,16	0,22
17	0.88±0.13	0,24			

spectrum of the laser radiation, just as its temporal spectrum, is continuous: the presence of a continuous part of the spectrum is due here to inhomogeneities of the radiating crystal. It is obvious that a correct analysis of the fluctuations of the second harmonic must, generally speaking, take into account both the temporal and the spatial non-monochromaticities of the fundamental radiation.

To investigate the second-harmonic generation it is convenient to represent the laser radiation in the form of a quasi-monochromatic signal (see also [18]):

$$\mathbf{E}_1 = \mathbf{e}_1\{\mathcal{A}_1(\alpha, \beta, t) \exp [i(\omega_1 t - \mathbf{k}_1(\alpha, \beta)\mathbf{r})] + \text{c.c.}\}, \quad (13)$$

where

$$\mathcal{A}_1(\alpha, \beta, t) = \sum_{m=1}^{N} \mathcal{A}_{1m}(\alpha, \beta, t) \exp\{i(\omega_{1m} - \omega_1)t\},$$
$$\mathcal{A}_{1m} = {}^1\!/_2 A_{1m}(\alpha, \beta, t)\, e^{i\varphi_{1m}(t)}; \quad (14)$$

$\omega_1 = (\omega_{11} + \omega_{1N})/2$ is the average frequency of the spectrum, and $\mathbf{k}_1(\alpha, \beta)$ is the wave number for this frequency.

Assume that the radiation (13) propagating in an anisotropic dispersive medium with weak nonlinearity of quadratic type, being an ordinary wave, excites in the medium an extraordinary second-harmonic wave. We seek the field in the nonlinear medium in the form

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{e}_1\mathcal{A}_1(\mu\mathbf{r}, \mu t)\, e^{i(\omega_1 t - \mathbf{k}_1\mathbf{r})} + \mathbf{e}_2\mathcal{A}_2(\mu\mathbf{r}, \mu t)\, e^{i(\omega_2 t - \mathbf{k}_2\mathbf{r})}. \quad (15)$$

In (15) the index 2 pertains to the second-harmonic wave, $w_2 = 2\omega_1$, and the parameter μ ($\mu \ll 1$) has an order of magnitude E/E_{at}, where E_{at} is the atomic field. Using the ordinary procedure for deriving the abbreviated equations ([21]), we get for the description of second-harmonic generation in a bounded beam, using the prescribed field approximation ($|\tilde{A}_1|^2 \gg |\tilde{A}|^2$) and neglecting diffraction,

$$\frac{\partial \tilde{A}_1}{\partial z} + \frac{1}{u_1}\frac{\partial \tilde{A}_1}{\partial t} = 0,$$

$$\frac{\partial \tilde{A}_2}{\partial z} + \rho\frac{\partial \tilde{A}_2}{\partial x} + \frac{1}{u_2}\frac{\partial \tilde{A}_2}{\partial t} = -i2\Gamma\tilde{A}_1^2(r, t)\, e^{-i\Delta\mathbf{r}}, \quad (16)$$

where

$$\Gamma = \frac{\pi\omega_2^2}{k_2 c^2 \cos \widehat{\mathbf{k}_2\mathbf{s}_2}}(\mathbf{e}_2\hat{\chi}^{2\omega}\mathbf{e}_1\mathbf{e}_1),$$

$$\Delta = \Delta(\alpha, \beta) = \mathbf{k}_2(\alpha, \beta) - 2\mathbf{k}_1(\alpha, \beta);$$

$\hat{\chi}$ is the nonlinear-polarizability tensor of the medium; $u_1 = u_1(\alpha, \beta)$ and $u_2 = u_2(\alpha, \beta)$ are the group velocities of the fundamental and harmonic waves. The term $\rho\,\partial\tilde{A}_2/\partial x$ characterizes the "drift" of the energy of the second harmonic, connected with the difference between the directions of the ray vectors of the fundamental wave $\mathbf{s}_1$ and the second harmonic $\mathbf{s}_2$, leading to the so-called aperture effect (see [19, 20]), ρ is the angle between the ray vectors $\mathbf{s}_1$ and $\mathbf{s}_2$, the anisotropy angle (in (16) it is assumed that $\rho \sim \mu$). The direction of the z axis coincides with the directions of the normal to the separation boundary between the linear and nonlinear media and the axis of the laser cavity ($\alpha = 0$). The x axis is perpendicular to the z axis and lies in the plane of the ray vectors.

The system (16) must be solved under the following boundary conditions

$$\mathcal{A}_1(x, y, z = 0, t) = \mathcal{A}_1(x(\alpha, \beta, L),$$
$$y(\alpha, \beta, L), t) = \mathcal{A}_1(\alpha, \beta, L, t),$$
$$\mathcal{A}_2(x, y, z = 0, t) = \mathcal{A}_2(\alpha, \beta, L, t) = 0. \quad (17)$$

We recall that L is the distance from the hypothetical phase center of laser radiation.

In a nonlinear anisotropic medium there exists a cone of directions for which the synchronism condition is satisfied ($\Delta = 0$). If the direction of the z axis coincides with one of the synchronism directions, then, by drawing the y axis perpendicular to the x and z axes and measuring the angle β from the y axis, we obtain at $z = 0$

$$x = \alpha L \sin \beta, \qquad y = \alpha L \cos \beta. \quad (18)$$

Owing to the small angular divergence 2ϑ of the fundamental radiation, we have in first order in ϑ:

$$\Delta\mathbf{r} = \Delta_z z, \quad \Delta_z = \Delta(\alpha, \beta) = G\alpha \sin \beta = Gx/L. \quad (19)$$

The constant G is determined by the anisotropy and by the dispersion properties of the medium.

A solution of (16) under conditions (17) and (19) for the amplitudes A_1 and A_2 leads to the expressions

$$\mathcal{A}_1 = \mathcal{A}_1(x, y, L, t - z/u_1),$$

$$\mathcal{A}_2(x, y, l, t) = -i2\Gamma \int_0^l \mathcal{A}_1^2(x - \rho(l - z), y, L, t - l/u_2 + \gamma z) \times \exp\left\{-i\frac{G}{L}z(x - \rho(l - z))\right\}dz, \quad (20)$$

where the parameter γ characterizes the deviation of the group velocities

$$\gamma = u_2^{-1} - u_1^{-1} = u_2^{-1}(\alpha, \beta) - u_1^{-1}(\alpha, \beta). \quad (21)$$

For the power density of the second harmonic p_2 we get from (20)

$$p_2(\alpha, \beta, l, t) = p_2(x, y, l, t) = 2\mathcal{A}_2(\mathbf{r}, t)\mathcal{A}_2^*(\mathbf{r}, t)$$
$$= 8\Gamma^2 \int_0^l\int \mathcal{A}_1^2(x - \rho(l - z_2), y, L, t - l/u_2 + \gamma z_2)$$
$$\times \mathcal{A}_1^{*2}(x - \rho(l - z_1), y, L, t - l/u_2 + \gamma z_1)$$
$$\times \exp\left\{-i\frac{G}{L}[z_2(x - \rho(l - z_2)) - z_1(x - (l - z_1)\rho)]\right\}$$
$$\times dz_2\, dz_1. \quad (22)$$

If the field of the fundamental radiation is a random Gaussian process, stationary in time and homogeneous and isotropic in space, with a correlation function,

$$B_1(s, \tau) = \mathrm{Re}(E_1(\mathbf{r} + \mathbf{s}, t)E_1^*(\mathbf{r}, t)) = R(s, \tau)\cos\omega_1\tau, \quad (23)$$

then in this case we obtain for the mean value $\bar{p}_2$ in the near field of the source

$$\bar{p}_2(x, y, l) = 16\Gamma^2 \int_0^{l/2} dz \int_0^{2z} R^2(\rho z_1, \gamma z_1) \times \cos\left[(x - \rho(l - 2z))\frac{G}{L}z_1\right]dz_1. \quad (24)$$

Using (22) and (24), let us consider separately the influence of the temporal and spatial non-monochromaticities of the fundamental radiation on the second-harmonic generation.

1. Influence of the Temporal Non-monochromaticity

We shall assume here that the anisotropy of the nonlinear medium can be neglected ($\rho \approx 0$), so that the harmonic-generation process is influenced only by the temporal non-monochromaticity. For the case of generation of a harmonic by means of a radiation having a continuous spectrum, it follows from (24) that if the correlation times τ_c of the fundamental radiation are close to the group-delay times between interacting waves, $|\gamma|l$, then p_2 depends on l, as in the case of monochromatic waves. When $\tau_c < |\gamma|l$, the average power density of the second harmonic $\bar{p}_2$ is proportional to l and does not depend on the phase deviation Δ_z. In the latter case the harmonic-generation process has the character of the nonlinear incoherent scattering, and the length

$$l_{coh}^{(t)} = \tau_c|\gamma|^{-1} \quad (25)$$

can be called the coherent length of nonlinear interaction. At lengths $l < l_{coh}^{(t)}$ the harmonic-generation process is coherent. This is also the condition for the applicability of the quasistatic approximation in the theory of nonlinear wave processes.

Let us consider now the generation of a harmonic by a discrete spectrum (multimode radiation). In this case, to take into account the influence of the dispersion, it is necessary to bear in mind that each mode of the second harmonic and of the fundamental radiation propagates with its own phase velocity. If we are interested in the average energy density $\bar{p}_{2n}$ of the n-th mode of a harmonic of frequency ω_{2n}, formed, for example, only from the m-th mode of the fundamental radiation of frequency $\omega_{1m}(\omega_{2n} = 2\omega_{1m})$, then $\bar{p}_{2n}$ $(2\omega_{1m}, x, y, l)$ will be determined by formula (24), where $R = R(0, \gamma z)$ is now the correlation function of the m-th mode. The coherent length for this process is equal to

$$l'^{(t)}_{coh} = |v_{2n}^{-1} - v_{1m}^{-1}|^{-1}\tau_{1m}, \quad (26)$$

where v are the velocities of the modes and τ_{1m} is the correlation time of the m-th mode.

In real conditions $\tau_{1m} > 10^{-8}$ sec, and therefore the coherent lengths $l_{coh}^{(t)}$, whose estimates are given in Table II, are much longer than the lengths l_{cr} of the employed nonlinear crystals, which usually amount to 2–3 cm. Thus, the generation of an individual second-harmonic mode by laser radiation can be considered in the quasi-statical approximation. Substituting (14) in (22), recognizing that $\tau_r \gg (\Delta\omega)^{-1}$, we obtain for the average second-harmonic power density (in the quasi-static approximation for each mode) a value

$$\bar{p}_2(x, y, l) = \frac{1}{2}\Gamma^2\left\{\sum_{m=1}^{N} A_{1m}^4(x, y, L)\frac{\sin^2[\Delta(m, m)l/2]}{[\Delta(m, m)l/2]^2}\right.$$
$$+ 4\sum_{j=m+1}^{N}\sum_{m=1}^{N-1} A_{1j}^2(x, y, L)A_{1m}^2(x, y, L)$$
$$\left.\times\frac{\sin^2[\Delta(j, m)l/2]}{[\Delta(j, m)l/2]^2}\right\}, \quad (27)$$

$$\Delta(j, m) = \Delta_z + (j + m - N - 1)\gamma\Delta\omega. \quad (28)$$

Table II. Values of the space scales characterizing the generation of the second harmonic by radiation from a ruby laser ($\lambda = 0.7\mu$) and neodymium-glass laser ($\lambda = 1.06\mu$) in a KDP crystal.

Laser	θ_0, rad	L, cm	$\lvert\gamma\rvert$, sec/cm	ρ, rad	G, cm^{-1}	$l'^{(t)}_{coh}$ cm	l_d, cm	l_a, cm	l_θ, cm
Ruby	$3.3\cdot10^{-3}$	200	$\sim10^{-12}$	$31\cdot10^{-3}$	$8.4\cdot10^{3}$	10^4	25	21	$7\cdot10^{-2}$
Neodymium	$1.1\cdot10^{-3}$	600	$\sim5\cdot10^{-14}$	$35\cdot10^{-3}$	$4.9\cdot10^{3}$	10^6	500	20	$36\cdot10^{-2}$

It follows from (27) that the dispersion of the nonlinear medium can be neglected at distances

$$l < \{(N-1)\Delta\omega|\gamma|\}^{-1} = l_d \tag{29}$$

when the synchronism condition is satisfied ($\Delta_z = 0$) or

$$l < \{(N-1)|\gamma|\Delta\omega + |\Delta_z|\}^{-1} = \{l_d^{-1} + |\Delta_z|\}^{-1} \tag{30}$$

if $\Delta_z \neq 0$.

Conditions (29) and (30) determine the length l over which the corrections connected with Δ_z and with the width of the spectrum $(N-1)\Delta\omega$ of the fundamental radiation are negligible. For radiation from ruby and neodymium lasers, the values of l_d in a KDP crystal are listed in Table II, which shows that in ordinary experiments in the synchronism direction $l_d > l_{cr}$. Therefore, in the experiments we have

$$\{l_d^{-1} + |\Delta_z|\}^{-1} \lesssim l_{cr} \quad \text{for} \quad |\Delta_z| \gg l_d^{-1}. \tag{31}$$

In this case, as seen from (27) and (28), the detunings $\Delta(j, m)$ can be set equal to an average phase detuning Δ_z, which is the one influencing the harmonic-generation process. The finite width $(N-1)\Delta\omega$ of the fundamental-radiation spectrum does not affect the power of the harmonic. In this connection, we can expect further simplification in the investigation of the second-harmonic generation by a discrete spectrum: the phase detunings Δ_z can be assumed the same for all modes, and the analysis of the generation process can be carried out in the quasi-static approximation for the entire spectrum of the fundamental radiation as a whole. The latter makes it possible to omit the time derivatives from (16) ($\partial\bar{A}_1/\partial t = \partial\bar{A}_2/\partial t = 0$).

2. Effects of Spatial Non-monochromaticity

Let us consider first the influence exerted on the generation of the second harmonic by the spatial incoherence of the fundamental radiation. (in this part of the section we assume that $l \ll l^{(t)}_{coh}$). We see from (24) that for distances $l < |\rho|^{-1} r_{coh}$ (r_{coh} is the radius of spatial coherence) we have $R(\rho l, 0) \approx R(0, 0)$, so that the power density of the harmonic $\bar{p}_2$ depends on l in the same manner as for a plane monochromatic wave. On the other hand, if $l > |\rho|^{-1} r_{coh}$, then in those cases when the presence of the phase deviation Δ_z is immaterial, we have $\bar{p}_2 \sim l$. In analogy with the coherent length $l^{(t)}_{coh}$ (25) we can introduce here a characteristic length $l^{(s)}_{coh}$:

$$l^{(s)}_{coh} = |\rho|^{-1} r_{coh}. \tag{32}$$

The spatial incoherence of radiation does not affect the generation of the harmonic when $l < l^{(s)}_{coh}$.

If the fundamental radiation is completely spatially coherent, the non-monochromaticity is determined by the finite angular divergence. Disregarding for the time being the multimode structure of the laser radiation, we obtain for the second-harmonic power P_2 generated by a diverging beam, substituting (14) in (22) and taking (5) and (18) into account,

$$P_2 = \int_0^{2\pi} d\beta \int_0^{\pi} p_2(\alpha, \beta, l)\,\alpha\,d\alpha = {}^1\!/_2\Gamma^2 A_{10}^4 \int_0^{2\pi} d\beta \int_0^{\pi} e^{-\alpha^2/\theta_0^2}\alpha\,d\alpha$$

$$\times \iint_0^{l} \exp\{-[\rho^2(z_1^2 + z_2^2) - 2\alpha L(z_1 + z_2)\sin\beta]/2L^2\theta_0^2\}$$

$$\times \exp\left\{-i\frac{G}{L}[(\alpha L\sin\beta + \rho l)(z_2 - z_1) + \rho(z_1^2 - z_2^2)]\right\}$$

$$\times dz_1\,dz_2. \tag{33}$$

For values of l such that $\rho l < L\theta_0$, expression (33) can be simplified to

$$P_2 = {}^1\!/_2\Gamma^2 A_{10}^4 F_2(\theta_0),$$

$$F_2(\theta_0) = l_2 \int_0^{2\pi} d\beta \int_0^{\pi} e^{-\alpha^2/\theta_0^2} \frac{\sin^2[{}^1\!/_2\alpha G\sin\beta]}{[{}^1\!/_2\alpha G\sin\beta]^2}. \tag{34}$$

The length

$$l_a = |\rho|^{-1} L\theta_0, \tag{35}$$

which depends on the anisotropy of the medium ρ and on the finite dimensions of the "spot" of fundamental radiation at the input of the nonlinear medium $L\theta_0$, is called the aperture length (see also [20]). An estimate of the values of l_a for radiations of ruby and neodymium lasers in a KDP crystal, given in Table II, yields $l_a > l_{cr}$. Consequently, we can use formula (34) to calculate the second-harmonic power.

The value of the function $F_2(\theta_0)$ in (34) depends on the relation between the length of the nonlinear crystal l_{cr} and the characteristic length[3)]

$$l_\theta = 2 / G\theta_0, \tag{36}$$

which is connected with the divergence of the fundamental radiation and with the constant of the medium G (see (19)). When $l > l_\theta$, the function $F_2(\theta_0) = \pi\theta_0^2 l^2$, and when $l > l_\theta$ (such cases are indeed realized in experiment, see Table II), we have

$$F_2(\theta_0) = 2\pi^{3/2}\theta_0 l G^{-1} = \pi^{3/2}\theta_0^2 l l_\theta. \tag{37}$$

4. CALCULATION OF THE "EXCESS" FLUCTUATIONS OF SECOND-HARMONIC POWER. QUASI-STATIC APPROXIMATION

The results of the theory developed in Sec. 3 show thus that to interpret the experimental data on the generation of harmonics from laser radiation we can confine ourselves to the quasi-static approximation and neglect the aperture effect. In this case the main sources of harmonic-power fluctuations will be the fluctuations of the amplitudes and the phases of the modes, the fluctuations in the number of the modes, and the fluctuations in the fundamental beam divergence.

For the second-harmonic current I_2 (Eq. (7b)), substituting (14) in (22) and taking into account (5), (18), and the condition $l < l_a$, we obtain in the quasi-static approximation the following value:

$$I_2 = a_2\Gamma^2 F_2(\theta_0) V_2(A_{10}(0,t), \varphi_{11}, \ldots, \varphi_{1N}). \tag{38}$$

Here $F_2(\theta_0)$ is determined by (37), $a_2 = a_{20}/2$, and

$$V_2 = A_{10}^4(0,t)\left\{N(2N-1) + 2\sum_{m,\, l=1}^{N}\cos\Phi_{nnml} + \sum_{p,\, q,\, m,\, l=1}^{N}\cos\Phi_{pqml}\right\}, \tag{39}$$

where the summation in the first case is for $m \neq l$ and $2n = m + l$, and in the second case for $p \neq q$, $m \neq l$, and $m \neq p$, but $p + q = m + l$;

$$\Phi_{pqml} = \Phi_{pqml}^{(c)} + \Phi_{pqml}^{(nc)};$$

$$\Phi_{pqml}^{(\,)} = \varphi_{1p}^{(\,)} + \varphi_{1q}^{(\,)} - \varphi_{1m}^{(\,)} - \varphi_{1l}^{(\,)}.$$

The value of the function V_2 begins to depend on the phases φ_{1m} when N = 3. In this case the amplitude of one mode of the second harmonic depends on the phases φ_{11}, φ_{12}, and φ_{13}: this mode is produced both as a result of doubling of frequency ω_{12} ($2\omega_{12} = \omega_{23}$), and as a result of the mixing of the frequencies ω_{11} and ω_{13} ($\omega_{11} + \omega_{13} = \omega_{23}$).[2, 18] With increasing N, the number of modes of the harmonic whose amplitudes depend on the phases of the modes of fundamental radiation increases.

Using (38) and (8), we can obtain the value of K in the expression (9):

$$K = aF(\theta_0) V_2(A_{10}(0,t), \varphi_{11}, \ldots, \varphi_{1N}) V_1^{-2}(A_{10}(0,t)), \tag{40}$$

where

$$a = a_2 a_1^{-2}\Gamma^2,\ F(\theta_0) = (2\sqrt{\pi} G\theta_0^3)^{-1} l. \tag{41}$$

The mean value $\overline{K}$ is

$$\overline{K} = a\int F(\theta_0) w(\theta_0)\, d\theta_0 \sum_N (2 - 1/N)\mathcal{P}(N). \tag{42}$$

For the relative fluctuations ξ of the coefficient K (see (11)) we obtain

$$\xi^2 + 1 = \int F^2(\theta_0) w(\theta_0)\, d\theta_0 \left[\int F(\theta_0) w(\theta_0)\, d\theta_0\right]^{-2} \times \sum_N \{(2-1/N)^2(1+\xi_\varphi^2(N))\}\,\mathcal{P}(N) \times \sum_N \left\{(2-1/N)\mathcal{P}(N)\right\}^{-2}. \tag{43}$$

Here $\xi_\varphi^2(N)$ represents the relative fluctuations of the coefficient K, connected with the random scatter of the values of the phases $\varphi_{1m}^{(nc)}$. The value of $\xi_\varphi^2(N)$ is

$$\xi_\varphi^2(N) = 8\left\{\sum C_{N-n}^2 + 2\sum C_{N-2n}^2\right\} \Big/ N^2(2N-1)^2;\quad N \geqslant 3. \tag{44}$$

Expression (44) can be reduced to the form

$$\xi_\varphi^2(N) = \frac{2}{3}\frac{(N-1)(4N^2 - 11N + 3)}{N^2(2N-1)^2},\ N \text{ odd}; \tag{45}$$

$$\xi_\varphi^2(N) = \frac{2}{3}\frac{(N-2)(4N-7)}{N(2N-1)^2},\ N \text{ even}. \tag{46}$$

Figure 2 shows a plot of $\xi_\varphi(N)$. The maximum value of $\xi_\varphi(N)$ is 0.25 for N = 5. At sufficiently large values of N, the function $\xi_\varphi(N)$ decreases like $(1.5N)^{-1/2}$. It must be noted that the values

3)More details on the characteristic length l_θ, which determines the efficiency of generation of harmonics by a diverging beam, is found in [22,23].

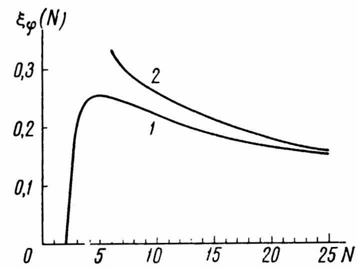

FIG. 2. Relative fluctuations $\xi_\phi(N)$ of the coefficients $K(I_2 = KI_1^2)$ due to the random scatter of phases in the fundamental radiation modes, vs. the number of modes of the fundamental radiation. Curve 1 corresponds to calculations according to the exact formula (44). Curve 2 corresponds to the asymptotic dependence $\xi_\phi(N) = (1.5\,N)^{-1/2}$.

of K and ξ do not depend on the fluctuations of the amplitudes A_{10} (see (43) and (42)).

In order to calculate the correlation coefficient R, it is necessary to know the quantities contained in (10), which are

$$\overline{I_1^2} = 8a_1^2A_0^4 \int F_1^2(\theta_0)\, w(\theta_0)\, d\theta_0 \sum_N N^2 \mathscr{P}(N),$$

$$\bar{I}_2 = 8a_2A_0^4 \int F_2(\theta_0)\, w(\theta_0)\, d\theta_0 \sum_N N(2N-1)\, \mathscr{P}(N),$$

$$\overline{I_1^4} = 384\, a_1^4A_0^8 \int F_1^4(\theta_0)\, w(\theta_0)\, d\theta_0 \sum_N N^4 \mathscr{P}(N),$$

$$\overline{I_2^2} = 384\, a_2^2A_0^8 \int F_2^2(\theta_0)\, w(\theta_0)\, d\theta_0$$

$$\times \sum_N \{1 + \xi_\varphi^2(N)\} N^2(2N-1)^2 \mathscr{P}(N),$$

$$\overline{I_2I_1^2} = 384\, a_1^2a_2A_0^8 \int F_2(\theta_0)\, F_1^2(\theta_0)\, w(\theta_0)\, d\theta_0$$

$$\times \sum_N N^3(2N-1)\, \mathscr{P}(N). \qquad (47)$$

In the derivation of (47) we took into account the laws governing the distributions of A_{10} and $w(A_{10})$ (Eq. (6)).

We assume first that $w(\theta_0) = \delta(\theta_0 - \overline{\theta_0})$ (δ—delta function), $\mathscr{P}(N_0)$ and $\mathscr{P}(N) = 0$ when $N \neq N_0$. Then the correlation coefficient (10) is equal to

$$R = \{1 + 1.2\xi_\varphi^2(N_0)\}^{-1/2} \qquad (48)$$

and has a minimum value of 0.97 at $N_0 = 5$. In this case R and ξ (Eq. (43)) are determined only by the phase fluctuations.

Knowing the distribution functions $w(\theta_0)$ and $\mathscr{P}(N)$, and using (43) and (47), we can calculate the values of R and ξ determined by the joint action of all the fluctuation sources. We assume that the distribution function of the angular divergence $w(\theta_0)$ is of the form

$$w(\theta_0) = [\theta_0'' - \theta_0']^{-1}$$
$$\text{for } \theta_0' \leqslant \theta_0 \leqslant \theta_0'', \qquad (49)$$

The distribution of $\mathscr{P}(N)$ is assumed to be binomial; for simplicity we represent it in the form

$$\mathscr{P}(N) = 2^{(1-N_m)} C_{N_m-1}^{N-1}. \qquad (50)$$

The calculated values of R and ξ are listed in Table III. The values of R_0 and ξ_0 were determined for a constant divergence 2ϑ, R_1 and ξ_1 for a divergence 2ϑ fluctuating in the interval from 20′ to 25′, and R and ξ_2 in the interval from 20′ to 30′ ($\theta_0 = 0.379\vartheta$ (5)); R and ξ were also calculated for three different values of N_m in the binomial distribution (50), namely $N_m = 2$, 4, and 7.

5. DISCUSSION

The experimental results of this paper show that in the generation of the second harmonic by means of a solid-state laser the correlation coefficient R between the harmonic current I_2 and the square of the fundamental current I_1^2 is in most cases smaller than unity and has values $R \approx 0.7$–1.0 (see Table I). The relative fluctuations of the coefficient K in the functions $A_2 = KI_1^2$, which were also determined experimentally, have essentially values 0.2–0.4 (see the same table).

The observed values of R and ξ (Table I) cannot be attributed solely to the presence of phase fluctuations at which relation (48) exists between R and ξ. Other factors affecting the values of R and ξ are the fluctuations of the beam divergence, the mode-number fluctuations, and the mode-amplitude fluctuations from spike to spike of the fundamental radiation. The theoretical (Table III) and experimental values of R and ξ point to a noticeable contribution made by these fluctuation sources.

The contribution of the fluctuation sources considered in this article changes if the coefficient

Table III. Theoretical values of the correlation coefficient R and of the relative fluctuations ξ of the coefficient K for different distribution functions of the number of modes $\mathscr{P}(N)$ and angular divergence $w(\theta_0)$

	$\mathscr{P}(N)$				$w(\theta_0)$
	1, $N=1$; 0, $N>1$	$^1/_2 C_1^{N-1}$	$2^{-3} C_3^{N-1}$	$2^{-6} C_6^{N-1}$	
R_0	1.00	1.00	0.91	0.98	$\delta(\theta_0 - \theta_0)$
ξ_0	0,00	0,20	0.22	0,24	
R_1	0.98	0.98	0,80	0.96	$w(\theta_0) = (1.90')^{-1}$
ξ_1	0,19	0.26	0.29	0.31	$7.58' \leqslant \theta_0 \leqslant 9.48'$ *
R_2	0.91	0.89	0,79	0.88	$w(\theta_0) = (3.79')^{-1}$
ξ_2	0,35	0.41	0,42	0.44	$7.58' \leqslant \theta_0 \leqslant 11.37'$ *

*The function $w(\theta_0)$ vanishes outside the indicated intervals of θ_0.

$\eta = P_2/P_1$ of transformation of the fundamental radiation into the second harmonic is large ($\eta \sim 1$). Thus, when $\eta \sim 1$ the phases $\varphi_{lm}^{(nc)}$ of the modes of the fundamental radiation in a nonlinear medium become correlated, and their one-dimensional distributions are altered. As a result, the contribution of the phase fluctuations to the "excess" fluctuations of power of the second harmonic becomes practically insignificant (see [15, 18]). There is likewise a decrease in the contribution from the fluctuations in the number of modes N and the mode amplitude fluctuations. Thus, for transformation coefficients $\eta \sim 1$, the main contribution to R and ξ is made by the fluctuations of the beam divergence.

In conclusion it must be emphasized that although, as shown above, the simplified quasi-static theory developed in Sec. 4 is adequate for an interpretation of the data obtained in experiments on harmonic generation with the aid of lasers, the results of several already published experimental papers can be explained only by using the more general theory developed in Sec. 3. We should mention here first experiments on second-harmonic generation with the aid of an ordinary (non-laser) light source,[24] and experiments on the mixing of coherent and incoherent light.[25] Spatial coherence effects should apparently play an essential role also in experiments on multiplication and mixing of frequencies of induced scattering lines (Raman or Mandel'shtam-Brillouin). In those cases when the induced scattering is obtained outside the optical resonator, its spatial coherence is apparently low. In all the foregoing cases the values of $l_{coh}^{(s)}$ will be small; the latter is apparently the main difficulty in attempts to realize accumulating nonlinear effects in the field of a non-laser source. The procedure developed here for calculating the fluctuation phenomena occurring during nonlinear optical interactions is, of course, applicable not only to problems involving the generation of harmonics and frequency mixing, but also to other nonlinear problems. Of particular interest may be a similar analysis applied to the theory of parametric interactions.

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105