STATISTICS OF PHOTOCOUNTS OF NONLINEARLY TRANSFORMED LIGHT

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Results are presented of a theoretical investigation of the statistical properties of the second optical harmonic produced in a homogeneous nonlinear crystal. Expressions are obtained for the intensity and count distribution of the harmonic excited by laser and thermal radiation. Quasistatic and nonstationary excitation conditions are considered. The effect of incomplete spatial coherence of the fundamental frequency on the statistical properties of the second harmonic is analyzed. It is found, in particular, that the temporal statistical properties of the harmonic may significantly depend on the spatial coherence of the fundamental radiation. In order to characterize the counting distribution, a parameter is introduced which is related only to the relative fluctuations of the radiation intensity.

INTRODUCTION

ALTHOUGH the nonlinear optical process most thoroughly investigated at the present time, both theoretically and experimentally, is generation of harmonics, the question of the influence of the statistical properties of the radiation on the character of the process itself, particularly on the maximum conversion coefficient, has not yet been sufficiently clarified. In the theoretical investigations performed to date, using various statistical models of the fundamental radiation, they studied either the average intensity and the variance of the second-harmonic intensity fluctuation[1-5], or else they calculated the coherence function in quadratic media[6-10]. The variance of the intensity of the fluctuations of a harmonic excited by laser radiation was investigated in[2-3], and the average intensity of the harmonic of thermal radiation was investigated in[11]. At the same time, knowledge of the laws governing the distribution of the intensity of the fundamental radiation and of the harmonic can yield both definite information concerning the nonlinear process and the properties of the nonlinear medium, and additional information concerning the statistics of the fundamental radiation.

In optics, the laws governing the distribution of the radiation intensities are determined by counting the number of electrons emitted by the photocathode of the detector that records the light beam[12-14]. A similar procedure is now being used extensively for the analysis of the statistics of laser radiation[15]. In experiments on nonlinear conversion of optical radiation, particularly on the generation of harmonics, this procedure can be used not only for the measurement of the statistics of the converted radiation[1)], but also for an accurate performance of experiments (for example, for an exact measurement of the nonlinear susceptibility tensor of a medium). Finally, nonlinear conversion of radiation with subsequent application of intensity interferometry (and in general amplitude interferometry) offers new possibilities and is a rather simple procedure for measuring the coherence functions of higher orders of the field of the fundamental radiation.

[1)]We note that a recent paper [16] considers the statistical properties of parametric luminescence.

The purpose of the present paper is a theoretical analysis of the one-dimensional laws of the distribution of the photoelectric counts of the second harmonic excited in homogeneous nonlinear crystals by gas-laser radiation and by non-laser thermal radiation. The analysis is based on a semiclassical description of the process of radiation registration. Indeed, by using gas lasers and gas-discharge sources it is easy to obtain in crystals of the KDP and $LiNbO_3$ type a second-harmonic intensity on the order of $1 \times (10^{-12}-10^{-10})$ W, corresponding to a harmonic photon density close to 10^6-10^8, and consequently the harmonic radiation can be described classically.

1. FUNDAMENTAL RELATIONS

In the semiclassical description of the process of registration of optical radiation, the distribution function of the probability of n photocounts is given by the expression (see[12,14])

$$P_m(n, T) = \int_0^\infty \frac{[\alpha_m U_m]^n}{n!} \exp\{-\alpha_m U_m\}\, w(U_m)\, dU_m, \tag{1}$$

where α_m is the quantum sensitivity of the detector, and U_m is the integral intensity during the observation time T:

$$U_m = U_m(T, t) = \int_t^{t+T} I_m(t')\, dt', \tag{2}$$

$w(U_m)$ is the intensity distribution function.

We shall henceforth assign to the index m the values 1 and 2, corresponding to quantities pertaining to the field of the fundamental radiation and of the second harmonic. To calculate the distribution of the photocounts of the fundamental radiation $P_1(n, T)$ and of the harmonic $P_2(n, T)$ it is therefore necessary to know respectively the distributions $w(U_1)$ and $w(U_2)$. The distribution of the intensity of the harmonic $w(U_2)$ can be found if information is available concerning the statistical properties of the fundamental radiation.

As is well known[2], the process of generation of the second harmonic by a wave having a temporal nonmonochromaticity at low transformation coefficients is described by abbreviated equations for the complex amplitudes $\tilde{A}_2$ and $\tilde{A}_1$ of the following form:

$$\frac{\partial \tilde{A}_2}{\partial z}+\frac{1}{u_2}\frac{\partial \tilde{A}_2}{\partial t}=-i2\gamma\tilde{A}_1^2,\quad \frac{\partial \tilde{A}_1}{\partial z}+\frac{1}{u_1}\frac{\partial \tilde{A}_1}{\partial t}=0 \tag{3}$$

with boundary conditions

$$\tilde{A}_2(t,z=0)=0,\quad \tilde{A}_1(t,z=0)=\tilde{A}_1(t).$$

In (3), the direction of the z axis coincides with the direction of the phase synchronism normal to the separation boundary of the linear and nonlinear media, u_m is the group velocity, and γ is the coefficient of nonlinear coupling. From (3) it is easy to find that

$$\tilde{A}_2(t,z)=-i\cdot 2\gamma\int_0^z A_1^2(t'+\nu\zeta)\,d\zeta, \tag{4}$$

where $t' = t - z/u_2$ and $\nu = 1/u_2 - u/u_1$ characterizes the difference between the group velocities. The connection between the instantaneous intensities of the fundamental radiation $I_1 = 2|\tilde{A}_1|^2$ and of the harmonic $I_2 = 2|\tilde{A}_2|^2$ in the so-called quasistatic approximation ($\nu = 0$) is algebraic:

$$I_2(t,z)=\Gamma I_1^2(t,z),\quad \Gamma=2\gamma^2z^2. \tag{5}$$

In the more general case (in the nonstationary regime of harmonic generation, $\nu \neq 0$), the relation between the intensities I_1 and I_2 is integral, and a dependence of I_2 on the fluctuations of the phases of the fundamental radiation appears:

$$I_2(t,z)=2\gamma^2\int_0^z\int_0^z I_1(t'+\nu\zeta_1)I_1(t'+\nu\zeta_2)\cdot \times\exp\{i\cdot 2[\varphi_1(t'+\nu\zeta_1)-\varphi_1(t'+\nu\zeta_2)]\}\,d\zeta_1\,d\zeta_2. \tag{6}$$

If the fundamental radiation has not only a temporal but also a spatial nonmonochromaticity, then allowance for the latter in the simplest approximation of geometrical optics leads to the relation (see[2])

$$I_2(t,x,y,z)=2\gamma^2\int_0^z\int_0^z I_1(t'+\nu\zeta_1,x'+\beta\zeta_1,y)I_1(t'+\nu\zeta_2,x'+\beta\zeta_2,y) \times\exp\{i\cdot 2[\varphi_1(t'+\nu\zeta_1,x'+\beta\zeta_1,y)-\varphi_1(t'+\nu\zeta_2,x'+\beta\zeta_2,y)]\}\,d\zeta_1\,d\zeta_2, \tag{7}$$

where $x' = x - \beta z$ and β = the birefringence angle, the angle between the wave vectors of the fundamental radiation and of the harmonic[2)].

We shall consider below, using expressions (1) and (4)–(7), the distribution of the intensity and of the photocounts of the second harmonic excited by laser radiation with statistically independent phases of the modes and thermal radiation with Gaussian statistics. Problems connected with the statistics of the harmonic from the laser radiation have been investigated, for simplicity, only in the quasistatic approximation; the influence of the nonstationary regime of generation and of the spatial coherence on the statistical properties of the harmonic is analyzed using as an example its excitation by thermal radiation.

2. DISTRIBUTION OF PHOTOCOUNTS OF THE SECOND HARMONIC EXCITED BY LASER RADIATION

In the general case, the radiation field of a laser operating in the free-generation regime has the following form[3)] at z = 0

$$E_1(t)=\sum_{k=1}^{K}\tilde{A}_{1k}(t)\exp\{i\omega_{1k}t\}+\text{c.c.}, \tag{8}$$

where K is the total number of modes; $\tilde{A}_{1k}$ and ω_{1k} are the amplitude and frequency of the k-th mode, $\omega_{1k} = \omega + (k - (K + 1)/2)\Delta\omega$, ω is the average radiation frequency, and $\Delta\omega$ is the frequency of the intermode beats. For subsequent analysis, expression (8) is conveniently written in the form

$$E_1(t)=\tilde{A}_1(t)e^{i\omega t}+\text{c.c.},$$

The integral intensity U_1 of the laser radiation in a time $T \gg (\Delta\omega)^{-1}$ is equal to

$$U_1=2T\sum_{k=1}^{K}|\tilde{A}_{1k}|^2=T\sum_{k=1}^{K}I_{1k}(t). \tag{9}$$

We also assume here that the period of the mode-intensity modulation is certainly longer than the time T. The expression for the intensity of the harmonic I_2, at an arbitrary number of modes K, is quite cumbersome even in the simplest case (5); we therefore proceed directly to a consideration of concrete problems.

Single-mode radiation. We begin the analysis with the case K = 1. We assume the fluctuations a of the amplitude A_1 to be small (in real notation)

$$A_1=A_0+a,\quad \sqrt{\overline{a^2}}/A_0=\mu\ll 1, \tag{10}$$

and to have a Gaussian distribution. For the distribution of the intensities of the fundamental radiation I_1 and of the harmonic I_2, generated in the quasistatic regime (5) (and consequently for U_1 and U_2), determined accurate to μ^2, we obviously again have a Gaussian distribution[4)]

$$w(I_m)=\frac{1}{\sqrt{2\pi}\,\sigma_m}\exp\left\{-\frac{(I_m-I_{m0})^2}{2\sigma_m^2}\right\}, \tag{11}$$

where $I_{10} = A_0^2/2$ and $I_{20} = 2\Gamma I_{10}^2$ are constant intensities and $\sigma_1^2 = 2\overline{a^2}I_{10}$ and $\sigma_2^2 = 32\overline{a^2}\Gamma^2 I_{10}^3$ are the variances of the fluctuations of the corresponding intensities. Thus, in the approximation under consideration, the intensities I_1 and I_2 have the same distribution. However, the relative fluctuations of the intensity $\xi_m = \sigma_m/I_{m0}$ are twice as large in the second harmonic as in the fundamental radiation.

The photocount distribution corresponding to the intensity distribution (11) is given by the expression

$$P_m(n)=\frac{(\sqrt{2}\,\alpha_m\sigma_mT)^n}{\sqrt{\pi}}H_{-(1+n)}\left[\frac{\alpha_m\sigma_mT-I_{m0}/\sigma_m}{\sqrt{2}}\right]\exp\left\{-\frac{I_{m0}^2}{2\sigma_m^2}\right\} \tag{12}$$

$H_n(x)$ are Hermite polynomials.

For small relative intensity fluctuations, the average number and the mean square of the number of photocounts are equal to

$$\bar{n}_m=\alpha_mTI_{m0},\quad \overline{n_m^2}=\bar{n}_m+\bar{n}_m^2[1+(\sigma_m/I_{m0})^2]. \tag{13}$$

From (13) at $\sigma_m \to 0$ we obtain the variance of the number of photocounts for a Poisson distribution. The excess of the fluctuations of the photocounts over the

2) We emphasize that in the present paper we analyze an interaction of the type 00–E.

3) For the questions considered here, the fact that the laser beams are bounded in space is of no fundamental significance.

4) Usually (see, for sample [17]), in theoretical calculations of the distribution of photocounts from a single-mode laser radiation one uses as a radiation model a random-phase harmonic signal plus noise. Then the intensity distribution is determined by the generalized Rayleigh distribution, but at low-intensity fluctuations (as is the situation with radiation from gas lasers) it goes over into a Gaussian distribution.

Poisson statistics is connected with the fluctuations of the intensity. It is possible to introduce a parameter d characterizing the difference between the distribution of the photocounts and the Poisson distribution:

$$d=\{\overline{n_m^2}-\bar{n}_m^2-\bar{n}_m\}^{1/2}/\bar{n}_m, \quad (14)$$

for the case (11) it is equal to

$$d=(\sigma_m/I_{m0})=\xi_m.$$

Expressions (12)—(14) are the general characteristics of the distribution of the photocounts for a Gaussian intensity distribution.

Thus, in the considered case of single-mode radiation, the distribution of the photocounts of both the fun fundamental radiation and of the second harmonic can be regarded as a Poisson distribution[5)], accurate to μ.

Averaging of expression (5) makes it possible to determine the value of the nonlinear coefficient γ in terms of the number of photocounts

$$\gamma=\left\{\frac{\alpha_1^2\bar{n}_2}{2\alpha_2 z^2(\overline{n_1^2}-\bar{n}_1^2)}\right\}^{1/2}.$$

The distribution of the photocounts (12), accurate to μ^2, is valid for multimode radiation of a gas laser and for the harmonic, when the number of modes in the fundamental radiation does not exceed two ($K \le 2$). On the other hand, if the number of modes is $K \ge 3$, then the statistics of the photocounts of the harmonic differs noticeably from the photostatistics of the fundamental radiation.

Three-mode emission. When K = 3, the integral intensity of the second harmonic is equal to (see[11])

$$U_2=C+B\cos(2\varphi_{12}-\varphi_{11}-\varphi_{13}),$$
$$C=\frac{1}{2}T\gamma^2z^2\left\{\sum_{j=1}^{3}A_{1j}^4+4\sum_{\substack{i,j=1\\(i>j)}}^{3}A_{1i}^2A_{1j}^2\right\}, \quad (15)$$
$$B=2T\gamma^2z^2A_{12}^2A_{11}A_{13},$$

φ_{ij} is the phase of the mode. It follows therefore that the main source of the fluctuations in U_2 is due to fluctuations of the phases of the fundamental radiation, whereby the depth of the random modulation of U_2 can reach almost 90%, which greatly exceeds the fluctuations of U_2 directly connected with the fluctuations of the amplitudes A_{1j}. The phases φ_{1j} in the free-generation regime can be regarded as statistically independent and as having a uniform distribution[18]. Then, assuming the amplitudes A_{1j} to be constant, we obtain for the distribution of the intensity U_2[19]

$$w(U_2)=1/\pi\sqrt{B^2-(U_2-C)^2},\qquad C-B\leqslant U_2\leqslant C+B. \quad (16)$$

The distribution of the photocounts, corresponding to the distribution (16), can be found with the aid of a generating function[14], which for the present case is of the form

$$Q(\lambda)=e^{-\lambda\alpha_2C}I_0(\lambda\alpha_2B). \quad (17)$$

Here $I_0(x)$ is the modified Bessel function and $0 \le \lambda \le 1$. The average number of counts and the variance of the number of counts are equal to

$$\bar{n}_2=\alpha_2C,\quad \overline{n_2^2}-(\bar{n}_2)^2=\bar{n}_2+{}^1/_2(\alpha_2B)^2, \quad (18)$$

and the parameter d introduced above (14) is equal to

$$d=\frac{1}{\sqrt{2}}\frac{B}{C} \quad (19)$$

and assumes a value d = 0.19 for different mode amplitudes A_{1j}.

Multimode radiation. With increasing number of modes of the fundamental radiation, the number of terms that depend on the phases of the modes in the expression for the integral intensity of the harmonic U_2 increases[2]. Then the problem of finding the distribution of the intensity U_2 reduces essentially to the problem of determining the distribution of a field (or an oscillation) with statistically independent modes. The resultant distribution of such a field tends rapidly to a Gaussian distribution, and the distribution of the photocounts consequently will satisfy expression (12). For the case of constant and equal amplitudes A_{1j}, the average intensity of the harmonic is $\overline{U}_{20} = 2(2 - 1/K)\gamma^2z^2TI_{10}^2$, and the relative fluctuations of the intensity U_2 are determined by the expression (see[2])

$$\xi^2=2(K-1)(4K^2-11K+3)/K^2(2K-1)^2,\quad K\text{—even},$$
$$\xi^2=2(K-2)(4K-7)/K(2K-1)^2,\quad K\text{—odd}, \quad (20)$$

($K \ge 3$). The dependence of the value of ξ on K is quite weak, $\xi = (1.5K)^{-1/2}$, and therefore the model of laser radiation with equal amplitudes is physically perfectly justified for the problem under consideration, especially at large K.

3. DISTRIBUTION OF THE PHOTOCOUNTS OF THE SECOND HARMONIC EXCITED BY THERMAL RADIATION

The distribution of the intensity due to thermal sources has an exponential form[12,14]

$$w(I_1)=I_0^{-1}\exp\{-I_1/I_0\}, \quad (21)$$

where I_0 is the average intensity. The photocounts of the thermal radiation are described by Bose-Einstein statistics:

$$P_1(n,T)=(1+\bar{n}_1)^{-1}(1+\bar{n}_1^{-1})^{-n}=(\bar{n}_1)^{-1}\exp\{-n/\bar{n}_1\}. \quad (22)$$

The last equation in (22) holds when $\bar{n}_1 \gg 1$, $\bar{n}_1 = \alpha_1TI_0$. For the case in question, we have d = 1 (Eq. (14)).

Quasistatic excitation regime. We disregard for the time being the partial spatial coherence of the thermal radiation, and consider first the generation of the harmonic by plane waves in the quasistationary regime (5). Then the distributions of the intensity and of the photocounts of the harmonic are given respectively by the expressions

$$w(I_2)=\frac{1}{2}\frac{\exp\{-\sqrt{I_2/\Gamma I_0^2}\}}{\sqrt{\Gamma I_0^2I_2}}$$
$$P_2(n)=\frac{(2n)!}{n!\,2^n(\bar{n}_2)^{1/2}}\exp\left\{\frac{1}{4\bar{n}_2}\right\}D_{-(1+2n)}\left(\frac{1}{\bar{n}_2^{1/2}}\right), \quad (23)$$

where $D_n(x)$ is the parabolic-cylinder function. The average number of photocounts is $\bar{n}_2 = 2\alpha_2T\Gamma I_0^2$ compared with the monochromatic fundamental radiation of the same intensity I_0 turns out to be twice as large. In this case $d = \sqrt{5}$. The distribution of the photocounts

5)A similar conclusion follows from [17], where a rigorous analysis has been carried out of the statistics of the photocounts of single-laser emission.

(23) differs significantly from both the Poisson distribution ($d = 0$) and from the Bose-Einstein distribution ($d = 1$).

Nonstationary excitation regime. In such a harmonic-generation process, the distribution of the photocounts of the harmonic tends to a Bose-Einstein distribution. Indeed, in a nonstationary regime (at distances $z \gg l_{coh}(t) = \tau_{coh}/|\nu|$, where τ_{coh} is the coherence time of the fundamental radiation) the statistics of the complex amplitude $\widetilde{A}_2$ (4) is Gaussian, in accordance with the central limit theorem. On the other hand, the distribution of the intensity of the harmonic (6) is exponential, with a mean value (see[4])

$$I_{20} = \bar{I}_2 = 8\gamma^2 z \int_0^\infty R^2(\nu\zeta)\, d\zeta,$$

where $R(\tau)$ is the coherence function of the complex amplitudes of the fundamental radiation.

The distribution of the photocounts of the harmonic can be noticeably influenced also by incomplete spatial coherence of the fundamental radiation. The statistics of the harmonic field becomes Gaussian as a result of the influence of the partial spatial coherence even in the quasistatic regime of generation, if the crystal length is $z \gg l_{coh}^{(sp)} = r_{coh}/\beta$ (r_{coh} is the radius of the spatial coherence of the fundamental radiation) (compare (16) with (7) at $\nu = 0$). In the general case, with incomplete space-time coherence of the fundamental radiation, the change of the statistics of the harmonic field, and consequently also of the photocount distribution, occurs over distances $z > \{(l_{coh}^{(t)})^{-2} + (l_{coh}^{(sp)})^{-2}\}^{-1/2}$.

CONCLUSION

An analysis of the distribution of the photocounts of the harmonic thus shows the following. In the case of the excitation of the harmonic by laser radiation with a small number of modes ($K \leq 2$), the statistics of the photocounts of the harmonics is close to Poisson statistics, and is analogous to the statistics of the photocounts of the laser radiation. When the number of laser modes $K \geq 3$, the distribution of the photocounts of the harmonic already differs noticeably from the Poisson distribution (the maximum value of the parameter $d \approx 0.25$). This is connected with the fact that in this case the integral intensity of the harmonic depends on the phase fluctuations in the modes of the fundamental radiation. An analysis of the distribution of the distribution of the photocounts of the second harmonic, excited by the multimode radiation of the gas laser, makes it possible to investigate correctly the contribution of the fluctuations of the phases of the fundamental radiation to the excess fluctuations of the intensities of the harmonic[6)], and it can be used for the investigation of the dynamics of phase synchronization. A recent investigation of the distribution of the photocounts of the second harmonic was carried out experimentally[20], and the results of the experiment were in satisfactory agreement with the theoretical predictions of Sec. 2.

Even more sensitive to the generation conditions is the distribution of the photocounts of the harmonic, excited by the thermal radiation. Here the parameter d, connected with the fluctuations of the intensity of the harmonic, has a value $d = 1$ in the nonstationary generation regime (the distribution function of the Bose-Einstein photocounts) and $d = \sqrt{5}$ in the quasistatic regime. The smoothing of the fluctuations of the harmonics in the nonstationary regime is due to interference effects. A similar phenomenon can result also from incomplete spatial coherence of the fundamental radiation, in the presence of which the "inertia" of the conversion is due to the birefringence of the nonlinear crystal.

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6)We note that when harmonics are generated by radiation of a solid-state laser there exists, besides the indicated source of excess fluctuations of intensity, also other sources (fluctuation in the number of modes, fluctuations in the angular divergence, etc.)[2].

[1] S. A. Akhmanov and A. S. Chirkin, Vestnik, Moscow State Univ. Phys. Ser. No. 5, 78 (1965).

[2] S. A. Akhmanov, A. I. Kovrigin, A. S. Chirkin, and O. N. Chunaev, Zh. Eksp. Teor. Fiz. 50, 829 (1966) [Sov. Phys.-JETP 23, 549 (1966)].

[3] J. Ducuing and N. Bloembergen, Phys. Rev. 133, A1493 (1964).

[4] S. A. Akhmanov and A. S. Chirkin, Radiotekhnika i élektronika 11, 1915 (1966).

[5] B. A. Sotskiĭ, Dokl. Akad. Nauk Beloruss. SSR 12, 888 (1968).

[6] M. Beran, J. Develis, and G. Parrent, Phys. Rev. 154, 1224 (1967).

[7] M. Beran and J. Develis, J. Opt. Soc. Amer. 57, 186 (1967).

[8] J. Zuckerman and J. Develis, J. Opt. Soc. Amer. 57, 1176 (1967).

[9] J. Zuckerman and J. Develis, J. Opt. Soc. Amer. 58, 175 (1968).

[10] H. Prakash, N. Chandra, and Vachaspati, Nuovo Cimento 57B, 161 (1968).

[11] D. H. MacMahon, J. Appl. Phys. 37, 4832 (1966).

[12] E. Wolf and L. Mandel, Rev. Mod. Phys. 37, 231 (1965).

[13] R. Glauber, Optical Coherence and Photon Statistics, in: Kvantovaya optika i kvantovaya radiofizika (Quantum Optics and Quantum Radiophysics) (translation anthology), Mir, 1966.

[14] J. Klauder and E. Sudarshan, Fundamentals of Quantum Optics, 4, N. Y., 1968.

[15] J. A. Armstrong and A. W. Smith, Progress in Optics, 1967, p. 212.

[16] B. Ya. Zel'dovich and D. N. Klyshko, ZhETF Pis. Red. 9, 69 (1969) [JETP Lett. 9, 40 (1969)].

[17] P. J. Magill and R. P. Soni, Phys. Rev. Lett. 16, 911 (1966).

[18] S. A. Akhmanov and A. S. Chirkin, Izv. vuzov Radiofizika 8, 579 (1965).

[19] B. R. Levin, Teoreticheskie osnovy statisticheskoĭ radiotekhniki (Theoretical Principles of Statistical Electronics), Sov. Radio, 1966.

[20] A. V. Grigor'ev, M. M. Popov, V. G. Tunkin, and A. S. Chirkin, Abstracts of paper at Fourth All-union Symposium on Nonlinear Optics, Moscow State Univ., 1968.

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