THE TWO-PHOTON TECHNIQUE FOR THE MEASUREMENT OF ULTRA-SHORT LIGHT PULSES AND THE EFFICIENCY OF NONLINEAR OPTICAL PHENOMENA

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Submitted July 31, 1970

Zh. Eksp. Teor. Fiz. 60, 1584–1592 (May, 1971)

The two-photon (TP) method is discussed which is widely used for investigating the structure of radiation from lasers operating in the synchronized mode regime. It is shown that for the analysis of the efficiency of two-quantum processes and the generation of the second optical harmonic it is quite sufficient to know the characteristics of the track of the TP fluorescence. In the case of incomplete synchronization of modes of the radiation the efficiency of nonlinear processes does not depend on the duration of the ultra-short pulses recorded by means of the TP method and is determined only by the value of the contrast coefficient R of the track. The estimates carried out explain the experimentally observed small gains in efficiency of nonlinear processes which occur in the field of radiations characterized by the value R = 2–2.5. In the paper it is also shown that the efficiency of a nonlinear process of arbitrary order can be determined by means of a set of correlation functions of intensity which depend on a single argument and the highest order of which corresponds to the order of the nonlinear process. A brief discussion is given of the efficiency of nonlinear processes of the third and fourth orders.

1. STATEMENT OF THE PROBLEM

FOR the analysis of the majority of nonlinear optical processes it is desirable to know the dependence of the intensities I of the interacting waves on the time. If we are interested only in the efficiency of a nonlinear process in the field of modulated radiation then, as a rule, it is sufficient to have information concerning the homogeneous law w(I) for the distribution of intensities I. Experimental data on the functions I(t) and w(I) can be obtained, however, only in some cases.

But if we are dealing with nonlinear phenomena in the field of radiation from a laser operating in the regime of mode synchronization, then the intensity of the laser radiation has fluctuations of duration $10^{-12}-10^{-13}$ sec. And at the present time the principal method of measuring the parameters of such ultra-short fluctuations consist of measuring the correlation functions of the intensity, in particular the autocorrelation function $G^{(2)}(\tau)$ (cf., for example, the reviews [1–3]). The function $G^{(2)}(\tau)$ can be measured with the aid either of the generation of the second harmonic or of the more widely used two-photon (TP) method.

The TP method of measurement includes the determination of the contrast R of the track of the TP fluorescence and of the time scale of the bright spot of fluorescence.[1–3] As a rule the problem of the interpretation of the measurements consists of drawing definite conclusions concerning the time structure of laser radiation. The value R = 1.5 corresponds to the case of nonsynchronized modes, while R = 3 corresponds to the case of completely synchronized modes. For lasers operating in the regime of mode synchronization the measured values of R in the majority of cases are contained in the range $2 < R < 3$, and this is usually associated with partial synchronization. In [4–5] for certain models of partial synchronization of modes intermediate values of the contrast coefficient R were calculated, $1.5 < R < 3.0$. But knowing from experiment only the lowest correlation function of intensity $G^{(2)}(\tau)$ it is not possible to give preference to some definite model of incomplete mode synchronization. Blount and Klauder[6] have shown that for this it is necessary to have at least information on the correlation functions for the intensity of second $G^{(2)}(\tau)$ and third $G^{(3)}(\tau_1, \tau_2)$ orders.

The object of the present work is to show that for the analysis of the efficiency of generation of the second harmonic and of two-quantum processes in the field of laser radiation with partially synchronized modes it is quite sufficient to know the characteristics of the track of the TP fluorescence. At the same time it turns out to be possible to study both the quasistatic regime of the generation of the second harmonic and in certain cases the nonstationary regime of doubling. But first we have considered a more general problem, viz., it is shown that the efficiency of a nonlinear process of arbitrary order can be calculated with the aid of a set of incomplete correlation functions of intensity the highest order of which corresponds to the order of the nonlinear process.

2. OPTICAL HARMONICS OF ARBITRARY ORDER AND MULTIQUANTUM PROCESSES

We consider nonlinear processes analytically described by the relation

$$I_k(t) = \Gamma_k I_1^k(t), \tag{1}$$

where $I_1(t)$ and $I_k(t)$ are the instantaneous intensities of laser radiation and of the nonlinear phenomenon being analyzed, k is the order of the latter, the coefficient Γ_k depends on the nature of the process.

In the absence of saturation effects expression (1) can be used to describe the generation of harmonics, multiquantum absorption and ionization, multiquantum detection, etc. Relation (1) describes nonlinear phenomena occurring in the so-called quasistatic regime.[7]

Experimentally one ordinarily measures quantities proportional to the energy

$$W_k = \int_0^{T_r} I_k(t)\,dt, \tag{2}$$

with the recording time[1)] T_r being much greater than the period of intermode beats T. If (1) is taken into account the measured quantities W_1 and W_k are related by the equation

$$W_k = \eta_k T_r^{1-k} \Gamma_k W_1^k, \tag{3}$$

where

$$\eta_k = T_r^{k-1} \left\{ \int_0^{T_r} I_1^k(t)\,dt \right\} \left\{ \int_0^{T_r} I_1(t)\,dt \right\}^{-k}. \tag{4}$$

The quantity η_k characterizes the efficiency in the nonlinear process of modulated radiation compared to unmodulated radiation of the same intensity.[2)] Using the Helder inequality one can show that $\eta_k \geq 1$, with $\eta_k = 1$ for radiation of constant intensity. In the case of thermal radiation, as is well known, $\eta_k = k!$. From (4) it can be seen that the value of η_k can be calculated from a knowledge of the time structure of the basic radiation, i.e., $I_1(t)$. We shall show below that for real cases the value of η_k can be found having limited information concerning laser radiation contained in the parameters of the correlation function of the intensity. We introduce the correlation function

$$G^{(k)}(\tau_1, \tau_2, \ldots, \tau_{k-1}) = \int_0^{T_r} I_1(t) I_1(t+\tau_1) \ldots I_1(t+\tau_{k-1})\,dt, \tag{5}$$

which, following Glauber,[10] is usually called the correlation function of the k-th order. We restrict ourselves to a consideration of the case of the basic radiation with fixed, but randomly distributed phases of the modes; then the intensity $I_1(t)$ represents a periodic function of period T. Taking the foregoing into account we obtain for the function (5) the relation[3)]

$$\int_0^T \ldots \int_0^T G^{(k)}(\tau_1, \ldots, \tau_{k-1})\,d\tau_1 \ldots d\tau_{k-1} = \int_0^{T_r} I_1(t)\,dt \left[\int_0^T I_1(t)\,dt \right]^{k-1}. \tag{6}$$

Substituting (5) and (6) into (4) we obtain

$$\eta_k = \frac{T}{\tau_k} \eta_{k-1}, \qquad \tau_k = \int_0^T \frac{G^{(k)}(\tau)}{G^{(k)}(0)}\,d\tau, \tag{7}$$

where τ_k is a characteristic time scale of the correlation function $G^{(k)}(\tau)$.

Repeated application of formula (7) leads to the expression

$$\eta_k = T^{k-1} \left\{ \prod_{j=2}^{k} \tau_j \right\}^{-1}. \tag{8}$$

Thus, it turns out that for the calculation of the efficiency of a nonlinear process of the k-th order it is quite sufficient to have a set of correlation functions $G^{(2)}(\tau)$, ... $G^{(k)}(\tau)$. Until now it has been generally assumed[13] that for this it is necessary to know the complete correlation function $G^{(k)}(\tau_1, \tau_2 \ldots, \tau_k)$ which, as is well known, contains considerably more information than the set of functions indicated above.

It should also be noted that experimentally the measurement of the function $G^{(k)}(\tau_1, \ldots, \tau_{k-1})$ is very complicated even for the function of the third order (k = 3), and therefore until now only the first experiment has been carried out[14] on the measurement of the function $G^{(3)}(\tau_1, \tau_2)$. At the same time for the measurement of the incomplete correlation function $G^{(k)}(\tau)$ one can utilize nonlinear processes of corresponding order.

Thus, the function $G^{(3)}(\tau)$ has been measured with the aid of the generation of the third harmonic[15] and of three-photon fluorescence.[14] In connection with the foregoing calculation of the efficiency of a nonlinear process with the aid of formula (8) is considerably more convenient than by utilizing the function (5).

For the analysis of a nonlinear phenomenon characterized by the relationship between the intensity of the signal I_s under investigation and the intensity of laser radiation which differs from the power law (1):

$$I_s(t) = H(I_1(t)),$$

the function $H(I_1)$ can be expanded in a Taylor series:

$$I_s(t) = \sum_n \alpha_n I_1^n(t) \tag{9}$$

and then one can utilize formulas (3), (8).

In order to obtain the best approximation the coefficient α_n in (9) can be calculated by means of the least squared error (cf., for example,[16]).

Since until now the vast majority of experiments has been carried out on the measurement of the correlation function $G^{(2)}(\tau)$ we give below a detailed analysis of the efficiency of nonlinear processes of the second order.

3. THE SECOND HARMONIC AND TWO-QUANTUM PROCESSES. QUASISTATIC REGIME OF EXCITATION

We demonstrate how one can obtain the value of the efficiency η_2 for nonlinear processes from experimental data on two-photon fluorescence (TPF). For a particular experimental arrangement (pulse collision) TPF yields the function[1-3]

$$F(\tau) = \alpha[G^{(2)}(0) + 2G^{(2)}(\tau)], \tag{10}$$

where α is an apparatus constant.

Taking (10) into account we obtain for the quantity η_2 in (8)[4)]

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1) For pulsed solid state lasers the time T_r in the majority of cases is the time of duration of the train of pulses.

2) In [8] the efficiency of the second harmonic η_2 was measured as a function of the order of the transverse mode of laser radiation, while in [9] an estimate of the duration of the pulses of laser radiation was made on the basis of the value of η_2.

3) For the case k = 2 relation (6) was utilized in references [11,12].

4) An expression similar to (11) for determining the effective length of pulses of laser radiation has been given in a recent paper [12] devoted to the problem of the accuracy of measuring the characteristics of a TPF track.

$$\eta_2 = 2T\left\{\int_{-T/2}^{T/2}[3f(\tau)-1]\,d\tau\right\}^{-1}; \qquad (11)$$

here $f(\tau) = F(\tau)/F(0)$.

Expression (11) is a formula for calculating the efficiency η_2 in terms of the shape of the TPF track which is not associated with the concept of contrast. The function $f(\tau)$ can be represented in the form

$$f(\tau) = R^{-1}[1+(R-1)b(\tau)]. \qquad (12)$$

Here $b(\tau)$ characterizes the shape of the bright spot of the TPF track, with $b(0) = 1$ and $b(T \gg \tau \gg \tau_c) = 0$, where τ_c is the correlation time for laser radiation, $R = f^{-1}(T \gg \tau \gg \tau_c)$ is the track contrast. We then have

$$\eta_2 = 2R[3-R+3(R-1)(\tau_f/T)]^{-1}. \qquad (13)$$

In (13) the time τ_f is the so-called integral scale for the bright spot of fluorescence

$$\tau_f = \int_{-T/2}^{T/2} b(\tau)\,d\tau. \qquad (14)$$

Expression (13) shows clearly that in the general case the efficiency of nonlinear processes of the second order depends on the contrast R of the track and on the size τ_f of the bright spot of TPF. At the same time from (13) it follows directly (since $\eta_2 \geq 1$) that the values of R lie in the range $1 \leq R \leq 3$. For radiation of constant intensity $R = 1$ and $\eta_2 = 1$; in a field of fluctuating intensity the value of η_2 increases with increasing R. The contrast coefficients $1 \leq R \leq 1.5$ correspond to the efficiencies $1 \leq \eta_2 \leq 2$. The simplest model of radiation for these values evidently represents a superposition of a monochromatic and a Gaussian radiation. As has been noted already, the case $1.5 < R < 3$ is associated with an incomplete synchronization of radiation modes.

The value of τ_f in expression (13) can be estimated using the well-known values of the efficiency for the cases of radiation with completely unsynchronized and synchronized modes:[2]

$$\eta_2(R=1{,}5) = 2 - 1/N, \quad \eta_2(R=3{,}0) \approx 2N/3, \qquad (15)$$

where N is the number of radiation modes. From here we obtain

$$\tau_f(R=1{,}5)/T = 1/2N, \quad \tau_\Phi(R=3{,}0)/T \approx 3/2N. \qquad (16)$$

Thus, for the same number of modes in the radiation spectrum of a laser in going over from unsynchronized modes to synchronized ones the value of the time τ_f in accordance with (14) differs only by a factor of three. As a rule, for lasers operating in the regime of mode synchronization, the number $N \approx 10^3$, so that $(\tau_f/T) \approx 10^3$.

From (13) it follows that in this case for $3 - R \gg 10^{-2}$, i.e., up to contrasts $R \leq 2.9$ the time dimension of the TPF bright spot is not significant and the efficiency of a nonlinear process is determined by the formula

$$\eta_2 = 2R/(3-R). \qquad (17)$$

Physically this circumstance is due to the fact that in the given case a small part of the energy of laser radiation is concentrated in intense ultra-short pulses recorded by means of the two-photon method; but the principal energy of radiation appears in the background (in "long" pulses of relatively low intensity).

Expression (17) enables us to interpret quantitatively the data of experiments carried out in the field of radiation of a laser with partially synchronized modes, and explains the comparatively small gain in the efficiency of a nonlinear phenomenon for the values $R = 2$–2.5 obtained in the majority of experiments. An analysis of the question of accuracy shows that for contrasts $R = 2$–2.5 which are usually measured with an accuracy of $\approx 10\%$ we have satisfactory accuracy ($\approx 50\%$) for the calculation of the value of η_2 in accordance with (17).

In the case under consideration the presence in the measured TPF track of a weak modulation of the track background can be attributed to the inaccuracy in the determination of a contrast R and one can consider the function $f(\tau)$ to be smooth. At the same time in virtue of the low value of the ratio τ_f/T the time τ_f can be estimated very roughly, for example to an order of magnitude.

Under the condition $3 - R \ll 10^{-2}$ the time dimension τ_f of the TPF bright spot is of particular importance, and for the value of the efficiency η_2 in (13) we obtain the result $\eta_2 = T/\tau_f$ which is valid for radiation with completely synchronized modes. In this case the contrast $R = 3$ must be measured with an accuracy of $\approx 10^{-3}$. Kuznetsova[11,12] has pointed out the necessity for such accuracy of measuring characteristics of the TPF track to prove the presence of single pulses in laser radiation.

We now compare the efficiency of generation of the second harmonic at the same energy W_1 of laser radiation operating in different regimes. We take the following values of the quantities R and T_r:[17] in the regime of Q-modulation the duration of generation is $T_r^{\text{nonsynch}} = 1.5 \times 10^{-8}$ sec, while the value R is $R = 1.5$, and in the regime of mode synchronization $R \approx 2.2$ and the duration of the train of ultra-short pulses is $T_r^{\text{synch}} = 10^{-7}$ sec. Utilizing (3) and (17) we obtain the ratio of the energies of the harmonics: $\eta_W = W_2^{\text{synch}}/W_2^{\text{nonsynch}} = 0.5$, and this agrees well with the experimental data[17] $\eta_W^{\text{exp}} \approx 0.3$ obtained using a KDP crystal.

An experimental comparison of the efficiency of excitation of TPF by radiation with different degrees of mode synchronization was carried out by Wang and Baardsen.[18] Under the conditions of their experiment in accordance with our formulas $\eta_W \approx 0.5$ and this is by an order of magnitude closer to the experimental value $\eta_W^{\text{exp}} \approx 8$ [18] than the estimate $\eta_W \approx 10^3$ obtained in [18] from the time dimension of the spot in the TP method, i.e., in accordance with the formula $\eta_2 = T/\tau_f$ which is valid for completely synchronized modes.

4. THE NONSTATIONARY REGIME FOR THE EXCITATION OF THE SECOND HARMONIC

Information on laser radiation obtained by the TP method can in some cases be sufficient also for the analysis of nonstationary nonlinear processes of the second order, in particular of the generation of a harmonic. Generally speaking, in the nonstationary regime of the generation of the second harmonic in addition to the information concerning the intensity of laser radiation $I_1(t)$ it is necessary to have information concerning the phase of the radiation $\varphi_1(t)$. In such a regime of generation the intensity of the second harmonic in the approximation of a given field is determined by the expression (cf., [19]).

$$I_2(t,z)=\Gamma_2 z^{-2}\int_0^z\int I_1(t+\nu x)I_1(t+\nu y)\times\exp\{i\cdot 2[\varphi_1(t+\nu x)-\varphi_1(t+\nu y)]\}\,dx\,dy, \quad (18)$$

where z is the length of the nonlinear crystal, $\nu = 1/u_2 - 1/u_1$ is the detuning of the reciprocal group velocities of the interacting waves. Thus, for the investigation of this regime it is necessary to know the combined statistical properties of $I_1(t)$ and $\varphi_1(t)$. For this one can utilize, for example, the quasistatic regime of frequency doubling (type of interaction OE → E) with subsequent amplitude interferometry of the radiation of the harmonic; however for solid state lasers such measurements so far have not been realized.

Therefore, here as an example of the excitation of a harmonic in a nonstationary regime[5)] we consider the model of basic radiation with periodic modulation of the intensity $I_1(t)$ and with fluctuations of the phase $\varphi_1(t)$ obeying the diffusion law.[6)] Carrying out a statistical averaging of expression (18) for the efficiency of excitation of the second harmonic in the case under consideration we obtain

$$\eta_2^{ns}=\eta_2/p. \quad (19)$$

Here η_2 is the quantity analyzed above while p has the form

$$p=z^2G^{(2)}(0)\left\{\int_0^z\int G^{(2)}[\nu(x-y)]\exp(-4D\nu|x-y|)\,dx\,dy\right\}^{-1}, \quad (20)$$

2D is the diffusion coefficient for the phase $\varphi_1(t)$. In terms of the characteristics of the TPF track the quantity p is given by the expression

$$p=2R\left\{(3-R)h(dz)+6(R-1)\int_0^z(z-\xi)b(\nu\xi)e^{-d\xi}\,d\xi\right\}^{-1}, \quad (21)$$

where d = 4Dν and

$$h(x)=2x^{-2}(e^{-x}+x-1). \quad (22)$$

Let the TPF bright spot be of exponential shape. Then for the coefficient η_2^{ns} in (19) we obtain, taking (13) into account

$$\eta_2^{ns}=\frac{(3-R)h(dz)+3(R-1)h[(d+s)z]}{3-R+3(R-1)(\tau_h/T\ln 2)}; \quad (23)$$

here $s = 2l_{qs}^{-1}\ln 2$, $l_{qs} = \tau_h/\nu$ is the quasistatic length,[7,20] τ_h is the time scale of the spot at the half-value level. For dz, sz ≪ 1, i.e., for lengths $z \ll l_{qs}$ and $z \ll l'_{qs} = 1/D\nu$, expression (23) reduces to the form (13).

In the nonstationary regime for the excitation of the harmonic ($z > l_k$ or $z > l'_k$), in contrast to the quasistatic regime the value of the coefficient η_2^{ns} depends significantly on the time dimension of the TPF bright spot for any arbitrary values of the contrast R. The efficiency of the harmonic falls with decreasing values of the characteristic lengths l_k and l'_k (h(x) → 0 with increasing x); in this case an important role can be played by phase modulation. Indeed, for contrast coefficients $R \approx 3$

$$\eta_2^{ns}(R\approx 3)\approx h[(d+s)z](T/\tau_h)\ln 2. \quad (24)$$

If $R < 3$ and $s \gg d$, then

$$\eta_2^{ns}(R<3)=h(dz) \quad (25)$$

then, consequently, $\eta_2^{ns} < 1$ for $dz \gtrsim 1$.

In the case of relatively weak phase fluctuations (dz ≪ 1) but of strong amplitude fluctuations (sz ≫ 1) we have

$$\eta_2^{ns}(R<3)=1,\quad \eta_2^{ns}(R\approx 3)=T/\nu z. \quad (26)$$

The quantity νz represents the time of group delay between interacting waves, in an essentially nonstationary regime it is equal to the duration of the pulse of the harmonic being excited[20] ($\nu z \gg \tau_h$). The values of η_2^{ns} determined by formulas (26) are related to the monochromatization of the spectrum of radiation of the second harmonic.[2,7]

In experiments[17,20] carried out in the field of laser radiation characterized by contrast $R \approx 1.8$–2.3 and of duration $\tau_h \approx 3$ nsec an efficiency was obtained for the excitation of the harmonic in the nonstationary regime in a crystal of $LiNbO_3$ (of length z = 1 cm, l_{qs} = 0.6 cm) which is by a factor of two or three greater than in the quasistatic regime in KDP (z = 2.5 cm, l_{qs} = 30 cm). An estimate in accordance with formulas (3), (23) for the conditions mentioned above, yields $\eta = W_2^{ns}/W_2 \approx 10$. The difference between this value and the experimental one can be due to the following causes: partial saturation of the transformation in the crystal of $LiNbO_3$ which has a greater nonlinear susceptibility and by the phase modulation of laser radiation. Although in [20] there was good agreement between the measured width of the spectrum and the duration of τ_h, this circumstance, however, does not exclude the possibility of phase modulation of the radiation with a spectrum width of the order of magnitude of the measured one. Taking the latter circumstance into account on the basis of the phase modulation model considered here can yield $\eta_2^{ns} \approx 4$.

CONCLUSION

The results of the present paper relating to the two-photon method of investigating the time structure of laser radiation can be generalized to the case of nonlinearly transformed radiation (for example, harmonics) and of the n-photon method. Such a generalization is of interest for the analysis of nonlinear phenomena of order higher than the second.

Thus, for the analysis of the efficiency of generation of the third harmonic and of three-quantum processes in addition to the quantity η_2 in (13) one must also know the value of $\eta_{3,1} = T/\tau_3$ (in accordance with (8), $\eta_3 = \eta_{3,1}\eta_2$), which if one utilizes the three-photon method analogous to the two-photon method discussed above, is determined by the expression

$$\eta_{3,1}=9R_3\{10-R_3+10(R_3-1)(\tau_f/T)\}^{-1}, \quad (27)$$

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5)The nonstationary regime of doubling of the frequency of radiation with completely synchronized modes and with linear frequency modulation has been investigated in [20,21].

6)With the aid of such a model of laser radiation one can explain, in particular, results of experiments on the compression of ultra-short light pulses [22].

where R_3 is the contrast of the track of three-photon fluorescence, excited by the laser radiation ($1 \leq R_3 \leq 10$).

The efficiency of nonlinear processes of the fourth order occurring in a field of radiation of modulated intensity can be conveniently determined with the aid of the formula

$$\eta_4 = \eta_{2,2}\eta_2^2, \tag{28}$$

where the quantity $\eta_{2,2}$ is analogous to η_2 but only is calculated in accordance with the data on the TPF excited by the second harmonic of laser radiation.

From (13), (27), and (28) it follows that the time dimensions of the bright spots of fluorescence play an important role only in the case of almost completely synchronized modes. In the case of partially synchonized modes—a regime in which in the majority of cases laser generators operate when modes are synchronized,—for the analysis of the efficiency of nonlinear processes it is quite sufficient to have information on the track contrast for multiphoton fluorescence. This particular feature is a characteristic property of the multiphoton method with colliding pulses.

The author is grateful to S. A. Akhmanov, R. Yu. Orlov, T. Usmanov, and R. V. Khokhlov for useful discussions of the present work.

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Translated by G. Volkoff
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