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KINETICS OF PARAMETRIC TRANSFORMATION OF INCOHERENT OPTICAL RADIATION

I. A. MARUSHKO and V. S. MASHKEVICH

Physics Institute, Ukrainian Academy of Sciences

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We investigated parametric transformation (amplification and generation) of light in nonlinear crystals. The quantum-transition method was used. The obtained kinetic equations make it possible to investigate the conditions for excitation and generation of light, to study the dependence of the efficiency of conversion on the pump-field radiation in the entire pump region. In a number of limiting cases this dependence is represented in very lucid form. In particular, it is shown that the conversion efficiency exhibits saturation at large pump values. The calculation results are applicable to the case of radiation that has no time coherence in the stationary or quasi-stationary generation regime.

1. INTRODUCTION

UNTIL recently, the theory of parametric transformation of light waves in nonlinear crystals was developed predominantly for the case of monochromatic ideally-coherent radiation[1-7]. By coherent state of the field we mean the eigenstates of the photon annihilation operator[8]. It is precisely in the case of such states that the classical nonlinear Maxwell's equations for the amplitudes of the field can be obtained from the equations of quantum electrodynamics by simply replacing the field operators with quantum-mechanical mean values of the operators. In the general case, the transition from the quantum Maxwell's equations to the classical ones is not obvious, even in the case of strong fields, without making special assumptions concerning the properties of the field.

In the case of stationary fields, as is well known[8], the condition of field coherence presupposes that the field is monochromatic. In this case, the finite width of the emission spectrum is to a certain degree a measure of the incoherence of the field. With the aid of the classical equations, written for coherent fields, it is obviously possible to investigate fields that are not ideally coherent, provided the deviations from the ideal character are small. Such a situation takes place, apparently, in the case of gas lasers. On the other hand, if these deviations are not small, then the solution of the nonlinear-optics problems holds for either derivation of corresponding classical Maxwell's equations from the quantum ones, and an investigation of their solution, or else the development of another convenient approximate method. One of such methods may be the method of quantum transitions (the kinetic method) developed and used for the investigation of generation of optical harmonics[9-10]. As will be seen from the following, the kinetic method is applicable to the case of essentially incoherent radiation. Of course, we are dealing here with the absence of coherence in the sense of[8]. Spatial coherence can take place. Such a situation can apparently be realized in the case of powerful solid-state lasers.

The availability of results of the wave and kinetic methods in the theory of radiation transformation, which pertain, strictly speaking, to different situations, makes it possible to investigate in detail coherent and statistical properties of the radiation of different sources. The convenience of using the kinetic method lies also in the fact that in this method it becomes possible to investigate the transformation of the radiation in all the intervals of values of the applied-field intensities, including the region of quantum noise in the converter[11,12], which is beyond the framework of the classical theories. The purpose of the present paper is to investigate the efficiency of parametric conversion of incoherent optical radiation by a kinetic method in the entire interval of values of the intensities of the electromagnetic field applied to the crystal.

2. THE METHOD OF KINETIC EQUATIONS

If we analyze the phenomenon of frequency conversion by the quantum-transition method, then we calculate the radiation intensity at any particular frequency by using the kinetic equations based on elementary considerations of the transition probabilities. The transition probabilities are calculated in accordance with the following scheme. We consider a certain converting device, containing a nonlinear element and placed in an electromagnetic field. The field is normalized at a certain volume, the dimensions of which are large compared with the dimensions of the apparatus. If we neglect the nonlinear properties of the system, then the normal oscillations of the field in this system will be waves satisfying the linear Maxwell's equations and the corresponding boundary conditions. The quantization of these waves leads to photons, understood to mean the occupation numbers of the indicated normal oscillations of the field. The spatial configuration of such a photon is determined by the linear dispersion properties of the apparatus. Account of the interaction of the field with the matter in the higher order of perturbation theory leads to interaction of the normal oscillations, i.e., to interaction between the photons. The probability of the process in which the interaction between the photons creates one photon in the j-th state with annihilation of an arbitrary pair of photons in the i-th and k-th states can be calculated in third-order perturbation theory by known methods [13]. In this case

$$W_j = \sum_{ik} B_{ikj}(q_j+1) q_i q_k \delta(\omega_i + \omega_k - \omega_j), \tag{1}$$

where in the dipole approximation

$$B_{ikj} = |\chi_{abc}^{ikj}|^2 \hbar (2\pi)^4 \omega_i \omega_k \omega_j F_{ikj}; \tag{2}$$

χ_{abc}^{ikj} is the tensor of the quadratic polarizability of the nonlinear medium;

$$F_{ikj} = \frac{1}{N^2(4\pi c^2)^3} \left| \sum_\alpha f_i(\mathbf{r}_\alpha) f_k(\mathbf{r}_\alpha) f_j^*(\mathbf{r}_\alpha) \right|^2, \tag{3}$$

N is the density of the atoms in the crystal, $f_i(r_\alpha)$ is the solution of the linear Maxwell's equation for the i-th state of the field, taken at the point where the α-th atom is situated; $q_{i,k,j}$ are the numbers of the photons in the corresponding states.

It is possible to calculate in similar fashion the probabilities of all the remaining processes that are conceivable in the three-photon interaction. The equations describing the time variation of the quantum-mechanical mean values of the photon numbers (the kinetic equations) can be written by starting from the expressions for the probabilities of these transitions and by introducing in suitable manner pumping of the states of the field and their losses. For the case when in three-particle interaction there is pumping at two frequencies, the kinetic equations take the form

$$\frac{dq_j}{dt} = \sum_{ik} B_{ikj}(q_j+1) q_i q_k \delta(\omega_i+\omega_k-\omega_j) - \sum_{ik} B_{ikj} \times q_j (q_i+1)(q_k+1)\delta(\omega_i+\omega_k-\omega_j) + N_j - \alpha_j q_j, \tag{4a}$$

$$\frac{dq_i}{dt} = \sum_{jk} B_{ikj} q_j (q_i+1)(q_k+1)\delta(\omega_i+\omega_k-\omega_j) - \sum_{jk} B_{ikj}(q_j+1) q_i q_k \delta(\omega_i+\omega_k-\omega_j) + N_i - \alpha_i q_i, \tag{4b}$$

$$\frac{dq_k}{dt} = \sum_{ij} B_{ikj} q_j (q_i+1)(q_k+1)\delta(\omega_i+\omega_k-\omega_j) - \sum_{ij} B_{ikj}(q_j+1) q_i q_k \delta(\omega_i+\omega_k-\omega_j) - \alpha_k q_k, \tag{4c}$$

where q must be taken to mean the average numbers of the photons in the states; N_j and J_i—pumping of the j-th and i-th states (number of photons entering per unit time); q—number of photons vanishing from the system per unit time as a result of all the processes not connected with the transitions in question, so that α—losses of the corresponding state or the reciprocal lifetime of the photon in the system. If there is no absorption in the system, then α—reciprocal photon lifetime relative to escape from the system, i.e., $\alpha = c/L$, where c—speed of light, and L—length of region of normalization of the field.

Equation (4) can be proved rigorously with the aid of equations for the density matrix of the matter plus a quantized radiation field system, if the following conditions are satisfied: (a) the regime is stationary, (b) perturbation theory is applicable, (c) the diagonal elements of the density matrix can be separated, i.e., the diagonal elements of the density matrix of higher order can be represented in the form of a product of diagonal elements of lower order.

Condition (a) makes it possible to exclude nondiagonal density-matrix elements, by equating the time derivatives of these elements to zero. If we forego the condition of strict stationarity, then we must require in place of condition (a) satisfaction of the following inequalities:

$$t > c/L, \tag{5a}$$

$$t\Delta\omega \gg 1, \tag{5b}$$

where t is the observation time (the duration of the radiation pulse) and $\Delta\omega$ is the spectral width of the radiation. Condition (5a) indicates that the stationary regime can be realized if the duration of the pulse exceeds the damping time of the natural oscillations of the system L/c. The arbitrariness of condition (5a) connected with the arbitrariness of the choice of L can be avoided by taking into account the fact that the definition of the normal oscillations holds true under the condition $L \gg l_0$, where l_0 is the length of the converting device. Therefore (5a) can be written in the form $t \gg l_0/c$. The latter is the condition for the establishment of stationary waves in the converting device, which are taken as the unperturbed states of the field. This condition is not connected with the character of the radiation conversion.

Condition (5b) can be regarded as the condition of nonmonochromaticity of the radiation, necessary for the existence of time-independent transition probabilities per unit time. In real pulsed solid-state lasers, this condition can be satisfied at least in those cases when no special selecting devices are used, to greatly narrow down the radiation spectrum.

Condition (c) is a definite assumption regarding the statistical properties of the field. It can be shown, using the results of [9], that this condition signifies the following connection between the square of the average number of photons $(\overline{q})^2$ and the mean square $(\overline{q^2})$ number of photons in the mode: $\overline{q^2} = 2(\overline{q})^2 + \overline{q}/2$. This condition is satisfied by radiation from a laser whose operation can be described by the kinetic method [14]. The statistics of such radiation is close to the statistic of incoherent radiation [14].

3. EFFICIENCY OF PARAMETRIC TRANSFORMATION

By parametric interaction we mean, in analogy with [4], an interaction in which only one of the several interacting frequencies has a powerful pump. We shall consider the case of transformation of frequency downward, putting $N_j \gg N_i$.

In the study of the interaction of spectral lines of finite width, interest attaches to the number of photons in the lines. From (4) we can go over to equations for the numbers of the photons in the lines, provided these lines are sufficiently narrow, so that within the limits of the line widths it is possible to neglect the dispersion of the quantity B_{ikj}. The condition of narrowness of the line width is not too stringent here. It is necessary only to simplify the calculations. For the KDP crystal in the transparency region, this condition is of the form $lc^{-1}\Delta\omega < 10^2$, so that when $l \approx 10$ cm we get $\Delta\omega < 10^{11}$ sec^{-1}. This condition can be satisfied in the case of laser sources. It is possible to find a region in which this condition does not contradict the condition (5b). For example, at a pulse duration $t = 10^{-7}$ sec we have, in accordance with (5b), $\Delta\omega \gg 10^{7}\ \text{sec}^{-1}$.

If we regard the propagation direction and the photon polarization in each line as fixed, then the summation over i, k, and j in (4) reduces to summation over the frequencies within the limits of the corresponding lines, which can be replaced by integration, introducing the density of the states ρ of the field with respect to the frequency. For plane waves $\rho = L/2\pi c$. The groups of states numbered by the indices j, i, and k will be denoted respectively by the indices p (pump), s (signal), and d (difference frequency). The numbers of photons in these groups (lines) are

$$q_p = \sum_j q_j, \quad q_s = \sum_i q_i, \quad q_d = \sum_k q_k.$$

Summing (4a)–(4c) respectively over j, i, and k under the indicated assumptions, we obtain

$$dq_p/dt = B[q_s q_d - q_p(q_s + q_d + M)] - \alpha_p q_p + N_p, \tag{6a}$$
$$dq_s/dt = B[q_p(q_s + q_d + M) - q_s q_d] + N_s - \alpha_s q_s, \tag{6b}$$
$$dq_d/dt = B[q_p(q_s + q_d + M) - q_s q_d] - \alpha_d q_d, \tag{6c}$$

where

$$N_p = \sum_j N_j, \quad N = \sum_i N_i, \quad M = \rho\Gamma, \quad B = B_{ikj}\rho$$

with i, k, and j taken for the centers of the corresponding lines: $\alpha_{s,p,d}$—losses at the central frequencies ω_s, ω_d, and ω_p ($\omega_p = \omega_s + \omega_d$); Γ—effective width of the spontaneous emission spectrum at the signal frequency. It is obviously defined as the maximum frequency interval in which B_{ikj} changes little. In the simplest case of the degenerate regime ($\omega_s = \omega_d = \omega_p/2$) and normal incidence of the light on the crystal, the estimate for Γ has the simple form:

$$\Gamma \approx \frac{2\pi c}{l\omega_p}\left[\frac{dn(\omega_p - \omega_i)}{d\omega_i}\bigg|_{\omega_i=\omega_s}\right]^{-1}, \tag{7}$$

where l is the length of the linear crystal and $n(\omega_p - \omega_i)$ is the refractive index. It is assumed, naturally, that the frequency of the applied signal field falls in the region of allowed spontaneous transitions. The system (6) in the stationary regime reduces to a quadratic equation for q_s, the root of which vanishes when $N_s \to 0$ and $N_p \to 0$, will be

$$q_s = (6B\alpha)^{-1}\{(2BN_p - \alpha^2)^2 + 12B\alpha[N_s(\alpha^2 - BN_p)\alpha^{-1} + BN_pM]\}^{1/2} + (2BN_p - \alpha^2)/6B\alpha. \tag{8}$$

We took into account here the fact that $N_s \ll N_p$, and we put $\alpha_s = \alpha_d = \alpha_p = \alpha$.

The radiation power at the frequencies ω_s, ω_d, and ω_p is

$$I_s = \hbar\omega_s\alpha q_s, \quad I_d = \hbar\omega_d\alpha q_d, \quad I_p = \hbar\omega_p\alpha q_p.$$

The signal gain coefficient $\mu = I_s/\hbar\omega_s N_s = \alpha q_s/N_s$. The conversion coefficient (or generator efficiency) is defined as $k = I_s/\hbar\omega_p N_p = \omega_s\alpha q_s/\omega_p N_p$. These coefficients are the system characteristics of interest to us, for a system operating in the parametric frequency conversion regime. Their calculation reduces to the use of (8). However, for practical calculations it is more convenient to use approximate but simpler expressions for q_s, obtained from (8) for a number of interesting regions of variation of N_p.

We note first that q_s differs from zero also when $N_s = 0$. This fact is connected with the quantum description of the field. In the case of weak pumps, satisfying the condition $2BN_p \ll \alpha^2$, we have for $N_s = 0$:

$$q_s = BN_pM/\alpha^2. \tag{9}$$

This radiation is connected with the spontaneous transitions, i.e., with the quantum noise of the converter[1)].

When $N_s > BN_pM\alpha^{-1}$, the quantum noise can be neglected and we obtain

$$q_s \approx \frac{N_s}{\alpha}\left(1 + \frac{BN_d}{\alpha^2}\right) \tag{10a}$$

when $2BN_p \ll \alpha^2$. Accordingly

$$\mu \approx 1 + BN_p/\alpha^2. \tag{10b}$$

Thus, we are dealing here with a weak amplification regime.

We represent the corresponding results for the following pump regions:

$$(\alpha^2 - 2BN_p)^2 \ll 12BN_s\alpha(\alpha^2 - BN_p), \tag{11a}$$
$$|BN_p - \alpha^2| \ll \alpha^2, \tag{11b}$$
$$BN_p \gg \alpha^2. \tag{11c}$$

We assume that the signal is sufficiently weak, so that the condition

$$3BN_s/\alpha^2 \ll 1 \tag{12}$$

is satisfied.

In the region (11a) we have

$$q_s \approx \sqrt{\frac{N_s}{6B}}\left(1 - \frac{2BN_p - \alpha^2}{\sqrt{6BN_s\alpha^2}}\right)$$
$$q_p \approx \frac{N_p}{\alpha}\left(1 - \sqrt{\frac{2BN_s}{3\alpha^2}}\right), \quad \mu \approx \sqrt{\frac{\alpha^2}{6BN_s}}. \tag{13}$$

From (12) and (13) we see that this is a region of appreciable gain, although the pump may be assumed as specified ($q_p \approx N_p/\alpha$).

In the region (11b) we have

$$q_s \approx \frac{N_p}{3\alpha}\left(1 - \frac{3BN_s(\alpha^2 - BN_p)}{\alpha^4}\right), \quad q_p \approx \frac{2N_p}{3\alpha},$$
$$\mu \approx \frac{1}{3}\frac{N_p}{N_s}, \quad k \approx \frac{\omega_s}{3\omega_p}\left(1 - \frac{3BN_s(\alpha^2 - BN_p)}{\alpha^4}\right) \tag{14}$$

Here we are dealing essentially with generation. We note that it is possible also when $N_s = 0$. The cause of the excitation of the generation, as seen from (4), is the spontaneous transitions. In the classical description of the field, the analog of the spontaneous transitions are the proposed fluctuations, in the presence of which excitation of generation becomes possible [4].

The region (11c) is the region of extremely strong pumping. Here

$$q_s \approx \frac{2N_p}{3\alpha}, \quad q_p \approx \frac{N_p}{3\alpha}, \quad k \approx \frac{2\omega_s}{3\omega_p}. \tag{15}$$

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1) During the course of revising this article, the authors learned that this radiation has been observed and called parametric luminescence [15].

Thus, the transformation coefficient reaches saturation.

An analysis of the degenerate regime entails no difficulty. To this end it is necessary to replace B_{ikj} in (4b) by $2B_{ikh}$, and Eq. (4c) can be discarded. We then obtain in the region of extremely strong pumping

$$k \approx \omega_s / \omega_p = 0.5. \quad (16)$$

Investigating in detail $q_s(N_p)$ and dq_s/dN_t, we can show that the generation of the subharmonics is a threshold process. The value of the threshold pump N_p lies between the regions (11a) and (11b). This makes it possible to present estimates for the threshold pumping:

$$\frac{\alpha^2}{2B} < (N_p)^{thr}_{gen} \lesssim \frac{\alpha^2}{B}. \quad (17)$$

The left side of this inequality is similar to the corresponding inequality obtained by the wave method [4]. Putting $(N_p)^{thr}_{gen} \approx \alpha^2/B$ and using (9), we write down an equation for the power of the spontaneous radiation in the case of weak pumping

$$I_s = BN_p M\hbar\omega_s\alpha^{-1} \approx \frac{N_p}{(N_p)^{thr}_{gen}} \frac{\hbar\omega_s\Gamma}{2\pi}. \quad (18)$$

This expression makes it possible to determine experimentally $(N_p)^{thr}_{gen}$ by measuring the width of the radiation spectrum and the power I_s as functions of the pump N_p without realization of the generation process itself.

The amplification process can also be a threshold process. It can be shown that if the weak-signal condition (12) is satisfied, then there exists a very narrow pump interval, in which the main part of the change vq_s/dN_p takes place when the pump is varied in the interval from $N \ll \alpha^2/2B$ to $N_p \lesssim \alpha^2/2B$. This fact is evidence of the threshold nature of the process. The threshold region is close to the region $N_p \approx \alpha^2/2B$. The latter makes it possible to estimate the threshold value of the pump for the amplification:

$$(N_p)^{thr}_{ampl} \approx \alpha^2/2B. \quad (19)$$

Of course, the signal should exceed the quantum noise in the system at these values of the pump.

In conclusion we note that the stationary solution of the system (6) is stable. This can be demonstrated in analogy with [16].

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