

# GENERATION OF PHOTON PAIRS AND FREQUENCY DOUBLING IN A MOVING REFERENCE FRAME

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Received 6 October 1995, revised 26 January 1996, accepted 31 January 1996

**Abstract.** A theory of the generation of pairs of photons with different frequencies by a strong plane wave which makes use of a moving reference frame is proposed. In this reference frame the excitation wave has the zero wave vector and, therefore, creates a purely time-dependent field. This allows one to apply the theory of quantum emission in a time-dependent medium and to give a nonperturbational description of the processes considered. Special attention is paid to the degenerate four-wave mixing when two identical photons generate two others, one of which is directed forward and the other backward. This process allows us to obtain frequency doubling in inverse-symmetrical media. Estimations show that it may be responsible for observations of frequency doubling in glass fibres and in gases.

**Key words:** nonlinear optics, quantum emission, Hawking process, four-wave mixing, down-conversion, frequency doubling.

## 1. INTRODUCTION

In this communication we consider nonlinear optical processes in which a strong planar wave causes the generation of pairs of photons with different frequencies. The simplest process of this type, called spontaneous photon decay, or parametric frequency down-conversion, transforms one initial photon into two final ones,  $1\omega_0 \rightarrow 1\omega_1 + 1\omega_2$ ;  $\omega_i$  being the frequency of a photon. Another process, called hyperparametric light scattering or degenerate four-wave mixing, which is also considered below, transforms two identical initial photons into two others:  $2\omega_0 \rightarrow 1\omega_1 + 1\omega_2$ . The first process is described by the second-order susceptibility  $\chi^{(2)}$ , the second one by the third-order susceptibility  $\chi^{(3)}$ . Both processes have a quantum origin; in the classical limit  $\hbar \rightarrow 0$  they disappear. (In the limit  $\hbar \rightarrow 0$  these processes take place only if the initial amplitude of at least one of the final waves differs from zero). These processes have already been considered in numerous earlier studies, see e.g. [1–4].

The goal of this paper is to propose a new approach to these processes, based on their consideration in a moving reference frame. We will show that this approach essentially simplifies the description, while the processes considered reduce to a simpler process of the generation of  $(\vec{k}', -\vec{k}')$  pairs of photons (superscript denotes the moving reference frame,  $\vec{k}'$  and  $-\vec{k}'$  being the wave vectors of generated photons). This allows one to obtain a nonperturbational solution of the problem considered. Besides, it also allows one to elucidate the analogy of the processes mentioned with such a fundamental quantum process as the black hole emission [5] and with an analogous emission recorded by an accelerating photodetector [6].

Here we pay the main attention to the degenerate four-wave mixing  $2\omega_0 \rightarrow 1\omega_1 + 1\omega_2$  when one of the generated photons ( $1\omega_1$ ) is directed forward and the other ( $1\omega_2$ ) backward. This process is of great interest as in the synchronization limit  $n_1 \rightarrow n_0$  it leads to frequency doubling in inverse-symmetrical media ( $2\omega_0 \rightarrow 1\omega_1$ ;  $n_i$  is the refractive index at  $\omega_i$ ). Note that frequency doubling has indeed been observed in glass fibres (see e.g. [7-12]) and gases [13-16]. Conventionally these observations are explained by breaking of inverse symmetry. We show here that the symmetry breaking mentioned is not indispensable for the occurrence of these effects.

The main idea of our consideration is the following: the process  $m\omega_0 \rightarrow 1\omega_1 + 1\omega_2$ ,  $m = 1, 2, \dots$  can take place only if  $n_{1(2)} > n_0$ . This allows one to introduce a moving reference frame (with the velocity  $v = c_1 n_0 / n_1 < c_1$ ) in which the initial wave has the zero wave vector and therefore generates a purely time-dependent perturbation of the mode  $\vec{k}_1$  ( $c_i = c/n_i$  is the phase velocity). In this reference frame, due to the momentum conservation law, the total momentum of generated photons is zero. It means that photons are generated by  $(\vec{k}'_1, -\vec{k}'_1)$  pairs. As a result, the original process  $m\omega_0 \rightarrow 1\omega_1 + 1\omega_2$  is reduced to the mixing of two,  $(\vec{k}', -\vec{k}')$ , modes in a time-dependent quantum medium.

The solution of this problem can be found by applying the theory of vacuum quantum effects in strong fields [6, 17]. According to this theory, a time-dependent perturbation of the zero-point state causes the generation of particles (photons). The underlying mechanism is due to the change of the physical content of field operators in time: the initial destruction operators become linear combinations of the final destruction and creation operators. Therefore, the state which is zeroth for the initial destruction operators is not that for the final destruction operators. This means that finally in the medium there appear photons. This mechanism of emission is analogous to the Hawking mechanism of black hole emission [5], according to which the gravitational acceleration causes the linear transformation of the field operators mentioned and, due to that, the emission of photons, see e.g. [5, 6] (for a discussion of the analogy mentioned see also [18-20]).

## 2. EQUATION FOR THE FIELD OPERATORS

Our task is to describe the emission of photons in a dielectric medium excited by a strong quasimonochromatic plane wave. The excitation wave is considered classically. In this approximation the excitation causes perturbation of initially nonexcited modes. The last modes, obviously, form an open quantum system in the time-space-dependent field. To describe the mode mentioned, one can use the equations of motion for the mode operators as they have the same form both for closed and open systems. A corresponding equation (e.g. Maxwell equation) for the mode  $\vec{k}$  reads [1]

$$\left( \frac{\partial^2}{\partial t^2} - c_k^2 \nabla^2 \right) \hat{\psi}_{\vec{k}} = -n_k^{-2} \frac{\partial^2}{\partial t^2} \hat{P}^{(nl)}. \quad (1)$$

Here  $n_k$  denotes the refractive index,  $c_k = c/n_k$ ,  $\nabla^2$  is the Laplace operator,  $\hat{P}^{(nl)}$  is the operator of nonlinear polarization. We suppose that  $\hat{P}^{(nl)} = \chi^{(2)} \hat{E}^2$  or  $\hat{P}^{(nl)} = \chi^{(3)} \hat{E}^3$ , depending on the process considered,  $\hat{E}$  is the operator of the strength of the electromagnetic field. The last operator contains two parts: the strong part  $E_0$  which describes the excitation and which can be considered classically, and the weak part  $\hat{\psi}$  which is the sum of the field operators of all quantum modes contributing to the equation for  $\hat{\psi}_{\vec{k}}$  ( $\hat{E} = E_0 + \hat{\psi}$ ).

Here we consider that  $E_0 = \mathcal{E}_0 \cos(\omega_0 t - \vec{k}_0 \vec{r})$ , where  $\mathcal{E}_0$  is the amplitude of excitation with slow time-space dependence. In this case Eq. (1) takes the form

$$\left( \frac{\partial^2}{\partial t^2} - c_k^2 \nabla^2 \right) \hat{\psi}_{\vec{k}}(t, \vec{r}) = -\frac{\partial^2}{\partial t^2} \eta(t, \vec{r}) \hat{\psi}(t, \vec{r}), \quad (1a)$$

where  $\eta(t, \vec{r}) = n_k^{-2} (m+1) \chi^{(m+1)} E_0^m(t, \vec{r})$ ,  $m = 1, 2$  (large term  $\sim E_0^{m+1}$  does not contribute to the solutions with the wave vector  $\vec{k}$ ). We suppose that  $c_k < c_0$  and that the excitation wave propagates along  $x$  direction:  $\vec{k}_0 \vec{r} = k_0 x$ . In this case  $E_0 = \mathcal{E}_0 \cos(\omega_0(t - x/c_0))$ ,

$$\eta(t, \vec{r}) = \eta_0 [m - 1 + \cos(m\omega_0(t - x/c_0))] \quad (2)$$

depends essentially only on  $t - x/c_0$ ;  $m = 1, 2$ ,  $\eta_0 = n_k^{-2} (1 + m^{-1}) \chi^{(m+1)} \mathcal{E}_0^m$ .

Let us introduce a moving reference frame  $(t', \vec{r}')$  with

$$t' = \gamma(t - x/c_0), \quad x' = \gamma(x - t\beta c), \quad y' = y, \quad z' = z, \quad (3)$$

where  $c \equiv c_k$ ,  $\gamma = (1 - \beta^2)^{-1/2}$ ,  $\beta = c/c_0 < 1$ . In this reference frame the initial wave has the zero momentum:  $E_0 = \mathcal{E}_0 \cos(\omega'_0 t')$ ,  $\omega'_0 = \omega_0/\gamma$ . Equation (1a) now takes the form

$$\left( \frac{\partial^2}{\partial t'^2} - c^2 \nabla'^2 \right) \hat{\psi}_{\vec{k}'}(t', \vec{r}') =$$

$$= - \left( \frac{\partial^2}{\partial t'^2} - 2\beta c \frac{\partial}{\partial t'} \frac{\partial}{\partial x'} + \beta^2 c^2 \frac{\partial^2}{\partial x'^2} \right) \bar{\eta}(t') \hat{\psi}(t', \vec{r}'), \quad (4)$$

where  $\bar{\eta} = \gamma^2 \eta(t/\gamma)$ ,  $k'_x = k' \cos \alpha'$ ,  $k' = k\gamma(1 - \beta \cos \alpha)$ ,  $\cos \alpha' = (\cos \alpha - \beta)/(1 - \beta \cos \alpha)$ ,  $\cos \alpha = k_x/k$ .

### 3. RELATION BETWEEN THE INITIAL AND FINAL OPERATORS

Solutions of Eq. (4) are plane waves:

$$\hat{\psi}(t', \vec{r}') = e^{i\vec{k}'\vec{r}'} \hat{\Phi}_{\vec{k}'}(t') + e^{-i\vec{k}'\vec{r}'} \hat{\Phi}_{-\vec{k}'}^+(t'), \quad (5)$$

where  $\hat{\Phi}_{\vec{k}'}(t')$  is a purely time-dependent linear operator

$$\hat{\Phi}_{\vec{k}'}(t') = \phi_{\vec{k}'}(t') \hat{a}_{\vec{k}'} + \xi \phi_{\vec{k}'}^*(t') \hat{a}_{-\vec{k}'}^+, \quad (6)$$

$\phi_{\vec{k}'}(t')$  is the  $c$ -function satisfying the differential equation

$$\left[ \frac{\partial^2}{\partial t'^2} (1 + \bar{\eta}(t')) + \omega'^2 (1 - \beta^2 \bar{\eta}(t') \cos^2 \alpha') - 2i\Gamma \frac{\partial}{\partial t'} \bar{\eta}(t') \right] \phi_{\vec{k}'}(t') = 0, \quad (7)$$

$\omega' \equiv \omega'_{\vec{k}'} = c k'$ ,  $\Gamma = \beta \omega' \cos \alpha'$ ,  $\xi$  is the constant which is determined below,  $\hat{a}_{\vec{k}'}^+$  and  $\hat{a}_{-\vec{k}'}^+$  are Bose operators. In (6) we take into account that the space dependence  $\exp(i\vec{k}'\vec{r}')$  corresponds to both  $\hat{a}_{\vec{k}'}^+$  and  $\hat{a}_{-\vec{k}'}^+$ .

Asymptotical solutions of Eq. (7) for  $\vec{k}' = \vec{k}'_1$  and  $t' \rightarrow \pm \infty$  (i.e. before and after excitation when  $\bar{\eta} = 0$ ) are linear combinations  $\mu_{\pm} \exp(-i\omega'_1 t') + \nu_{\pm} \exp(i\omega'_2 t')$ ; the frequency  $\omega'_2$  is determined by the equality  $\vec{k}'_2 = -\vec{k}'_1$ ; here  $\omega'_2 \geq 0$ .

For the following it is most important that  $\mu_+$ ,  $\nu_+$  differ from  $\mu_-$ ,  $\nu_-$ . For example, if one chooses  $\mu_- = 1$ ,  $\nu_- = 0$ , which corresponds to  $\hat{a}_{\vec{k}'_1}$  being the destruction operator before excitation, then  $\mu \equiv \mu_+ \neq 1$  and  $\nu \equiv \nu_+ \neq 0$ :

$$\phi_{\vec{k}'_1}(t') = \begin{cases} e^{-i\omega'_1 t'}, & t' \rightarrow -\infty, \\ \mu e^{-i\omega'_1 t'} + \nu e^{i\omega'_2 t'}, & t' \rightarrow \infty. \end{cases} \quad (8a)$$

$$(8b)$$

Actual values of  $\mu$  and  $\nu$  are determined by the asymptotical solution of differential equation (7) with initial condition (8a).

Substituting (8) into (6), one gets

$$\hat{\Phi}_{\vec{k}'_1}(t') = \begin{cases} e^{-i\omega'_1 t'} \hat{a}_{\vec{k}'_1} + \xi e^{i\omega'_2 t'} \hat{a}_{-\vec{k}'_1}^+, & t' \rightarrow -\infty, \\ e^{-i\omega'_1 t'} \hat{b}_{\vec{k}'_1} + \xi e^{i\omega'_2 t'} \hat{b}_{-\vec{k}'_1}^+, & t' \rightarrow \infty, \end{cases} \quad (9)$$

where  $\xi = \pm 1$  (due to independence of the commutator  $[\hat{\Phi}_{\vec{k}'_1}(t'), \hat{\Phi}_{\vec{k}'_1}^\dagger(t')]$  on time) and

$$\hat{b}_{\vec{k}'_1} = \mu \hat{a}_{\vec{k}'_1} + \xi \nu \hat{a}_{-\vec{k}'_1}. \quad (10)$$

Here  $\mu \equiv \mu_{\vec{k}'_1}$ ,  $\nu \equiv \nu_{\vec{k}'_1}$ ,  $|\mu|^2 - |\nu|^2 = 1$  (which follows from the equality  $[\hat{\Phi}_{\vec{k}'_1}(t', \xi = -1), \hat{\Phi}_{\vec{k}'_1}(t', \xi = 1)] = 2$ ). According to the time dependence (9),  $\hat{a}_{\vec{k}'_1}$  is the initial destruction operator, while  $\hat{b}_{\vec{k}'_1}$  is the final destruction operator.

Relation (10) between the initial and the final operator is analogous to the corresponding relation in the Unruh description of the accelerating photodetector [6, 21]. This relation allows one to express the intensity of the emission via  $|\nu_{\vec{k}'_1}|^2$ . Indeed, the empty photon state  $|0_i\rangle$  at  $t' \rightarrow -\infty$  ( $\hat{a}_{\vec{k}'_1}|0_i\rangle = 0$  for all  $\vec{k}'_1$ ), will contain at  $t' \rightarrow \infty$

$$N_{\vec{k}'_1} = \langle 0_i | \hat{b}_{\vec{k}'_1}^\dagger \hat{b}_{\vec{k}'_1} | 0_i \rangle = |\nu_{\vec{k}'_1}|^2$$

photons with the wave vector  $\vec{k}'_1$ . Photons are generated by  $(\vec{k}'_1, -\vec{k}'_1)$  pairs. This follows immediately from relation (10) which can also be presented in the form of the unitary transformation

$$\hat{b}_{\vec{k}'_1} = e^{\hat{S}} \hat{a}_{\vec{k}'_1} e^{-\hat{S}}, \quad (10a)$$

where

$$\hat{S} = \rho (\hat{a}_{\vec{k}'_1} \hat{a}_{-\vec{k}'_1} - \hat{a}_{\vec{k}'_1}^\dagger \hat{a}_{-\vec{k}'_1}^\dagger) = \rho (\hat{b}_{\vec{k}'_1} \hat{b}_{-\vec{k}'_1} - \hat{b}_{\vec{k}'_1}^\dagger \hat{b}_{-\vec{k}'_1}^\dagger),$$

$\rho = \xi \text{arch}|\mu|$ . According to this relation the initial and the final (f) zero-point state are related as follows:  $|0_i\rangle = \exp(\hat{S})|0_f\rangle$ . This means that in the state  $|0_i\rangle$  there are pairs  $(\vec{k}'_1, -\vec{k}'_1)$  of final photons.

#### 4. PARAMETRIC AND HYPERPARAMETRIC PROCESSES

Let us now return to Eq. (7). Taking into account expression (2) and restricting ourselves to terms up to the first order with respect to  $\eta_0$ , this equation transforms to

$$\left[ \frac{\partial^2}{\partial t'^2} + \bar{\omega}^2 (1 - h \cos \Omega'_0 t' + i g \sin \Omega'_0 t') \right] \bar{\varphi}_{\vec{k}'_1}(t') = 0, \quad (11)$$

where  $h = \eta_0 \gamma^2 (1 + \beta^2 \cos^2 \alpha')$ ,  $g = \eta_0 \gamma^2 (\Omega'_0 / \bar{\omega}) \beta \cos \alpha'$ ,  $\Omega'_0 = m \omega_0 \gamma^{-1}$ ,  $\bar{\omega} = \omega'(1 - mh/2)$ ,  $\bar{\varphi}_{\vec{k}'_1} = \phi_{\vec{k}'_1} \exp(\delta)$ ,  $\delta = \tilde{\eta} - i \omega' \beta \bar{\eta} \cos \alpha'$ . Introducing

the imaginary time shift  $t'_1 - t' = i(1/2\Omega'_0) \ln(h+g)/(h-g)$  and the dimensionless time  $z = \Omega'_0 t'_1/2$ , one obtains finally the Mathieu equation

$$\left( \frac{\partial^2}{\partial z^2} + a - 2q \cos 2z \right) \bar{\varphi}_{\vec{k}'_1} = 0, \quad (11a)$$

where  $a = (2\bar{\omega}/\Omega'_0)^2$ ,  $q = \bar{h}a/2$ ,  $\bar{h} = (h^2 - g^2)^{1/2}$ . This equation describes the classical oscillator with periodical time-dependent frequency. Here, as it is well known (see e.g. [17, 22]), the parametric resonance takes place. Solutions of Eq. (11a) with  $a = l^2$ ,  $l = 1, 2, 3, \dots$  grow exponentially with time. In the considered first approximation with respect to  $\eta_0$  only  $l = 1$  resonance should be accounted for. A corresponding solution of Eq. (11a) reads [17, 22]

$$\bar{\varphi}_{\vec{k}'_1}(t') = Ae^{st'_1} \cos(\Omega'_0 t'_1) + Be^{-st'_1} \sin(\Omega'_0 t'_1), \quad (12)$$

where  $s^2 = ((\bar{h}\bar{\omega})^2 - \epsilon^2)/16$ ,  $\epsilon = \Omega'_0 - 2\bar{\omega}$ , the constants  $A$  and  $B$  are determined by the initial conditions. This solution describes exponential (including very large) growth of the oscillator amplitude and therefore is essentially nonperturbational.

Supposing that the excitation is applied between  $t' = 0$  and  $t' = t'_0$ , and that it is adiabatically switched on and off, the initial "vibration"  $\bar{\varphi}_{\vec{k}'_1}(t') = \exp(-i\omega'_1 t')$ ,  $t' < 0$  transforms to the sum

$$\bar{\varphi}_{\vec{k}'_1}(t') = ch(st'_0)e^{-i\omega'_1 t'} + sh(st'_0)e^{i\omega'_2 t'}, \quad t' > t'_0. \quad (13)$$

Here we neglected the small imaginary time shift  $t'_1 - t'$  and took into account that under the adiabatic switch-off mentioned the positive frequency wave  $\exp(i\vec{k}'_1 \vec{r}'_1 - i\Omega'_0 t')$  transforms to the wave  $\exp(i\vec{k}'_1 \vec{r}'_1 - i\omega'_1 t')$ , while the negative frequency wave  $\exp(-i\vec{k}'_2 \vec{r}'_1 + i\Omega'_0 t')$  transforms to the wave  $\exp(-i\vec{k}'_2 \vec{r}'_1 + i\omega'_2 t')$ , and that  $\vec{k}'_2 = -\vec{k}'_1$ ,  $\omega'_2 \approx \Omega'_0 - \omega'_1$ . An analogous but reversed transformation takes place under the adiabatic switch-on mentioned.

A comparison of (13) and (8b) gives  $|\nu| = sh(st'_0)$ . The excitation duration  $t'_0$  in the moving reference frame can be found, e.g. if one considers the Gaussian excitation pulse with the characteristic duration  $t_0$ :  $\mathcal{E}_0 = \bar{\mathcal{E}} \exp[-(t - x/v_g)^2/4t_0^2]$ , where  $v_g$  is the group velocity at  $\omega_0$ , and  $\bar{\mathcal{E}}$  is the maximal amplitude of the excitation. In the moving reference frame  $\mathcal{E} = \bar{\mathcal{E}} \exp[-(t' - x'(c_0 v_g)/(c_0 v_g - c_1^2))/4t_0^2]$ , where  $t'_0 = t_0(\gamma|1 - c_1^2/c_0 v_g|)^{-1}$ .

Now the number of generated photons can be expressed as follows:

$$N_{\vec{k}'_1} = N_{-\vec{k}'_1} = shQ,$$

where

$$Q = 4^{-1} t'_0 \gamma^2 \omega'_1 (1 + \beta^2 \cos^2 \alpha'_1) [\eta_0^2 - (4(1 - \omega'_0/\omega'_1)/\gamma^2 (1 + \beta^2 \cos^2 \alpha'_1))^2]^{1/2}.$$

This number may be very large even for small  $|\eta_0|$  considered here, which means that the obtained solution is nonperturbational. One can also see that the processes under consideration have a threshold.

Let us return now to the laboratory reference frame. Then  $\vec{k}'_1$  transforms to  $\vec{k}_1$  and  $\vec{k}'_2 = -\vec{k}'_1$  to  $\vec{k}_2$ . Consequently, as it should be, the plane wave generates  $(\vec{k}_1, \vec{k}_2)$  pairs of photons. The number of generated photon pairs equals

$$N_{\vec{k}_1} = sh(Q_0 \xi [1 - (4(1 - \beta \cos \alpha_1 - m\omega_0/2\gamma^2\omega_1)/\xi\eta_0)^2]^{1/2}),$$

where

$$Q_0 = t_0\omega_1\eta_0/4|1 - c_1^2/c_0v_g|,$$

$$\xi = \gamma^2(1 - \beta \cos \alpha_1)[1 + \beta^2((\cos \alpha_1 - \beta)/(1 - \beta \cos \alpha_1))^2].$$

At that, due to the energy and momentum conservation laws,

$$\omega_1 \simeq m\omega_0(n_0 - n_2 \cos \alpha_2)/(n_1 \cos \alpha_1 - n_2 \cos \alpha_2),$$

$$n_2 \sin \alpha_2/(n_0 - n_2 \cos \alpha_2) = n_1 \sin \alpha_1/n_1 \cos \alpha_1 - n_0.$$

The angular distribution of the generated photons  $\vec{k}_1$  is  $N(\alpha_1) = sh(Q(\alpha_1))\rho(\alpha_1)$ , where  $\rho(\alpha_1) = d \cos \alpha'_1/d \cos \alpha_1 = (\gamma(1 - \beta \cos \alpha_1))^{-2}$ , the spectral distribution of the number mentioned is  $N(\alpha_1(\omega_1))d\alpha_1/d\omega_1$ .

## 5. FREQUENCY DOUBLING BY FORWARD-BACKWARD HYPERPARAMETRIC LIGHT SCATTERING

As an application of the presented theory we consider a degenerate four-wave mixing  $2\omega_0 \rightarrow 1\omega_1 + 1\omega_2$  when one of the generated photons ( $1\omega_1$ ) is directed forward and the other ( $1\omega_2$ ) backward. (Note that an analogous forward-backward process  $1\omega_0 \rightarrow 1\omega_1 + 1\omega_2$  has been considered in [1, 23] and observed in [24]). In this case the angles  $|\alpha_1|$  and  $|\alpha_2 - \pi|$  are small. Thereby  $\alpha_2 - \pi \approx \alpha_1 n_1(n_0 + n_2)/n_2(n_1 - n_0)$  and for  $|\eta_0| > |n_2 - n_0|(n_1 - n_0)^2 n_0^{-3}$

$$Q(\alpha_1) \approx Q_0(1 - 2\alpha_1^4(n_2 - n_0)^2/\eta_0^2 n_2^2), \quad (14)$$

where  $Q_0 = t_0\omega_0\eta_0/2|1 - c_1^2/c_0v_g|$ ,  $\eta_0 = 2\mathcal{E}_0^2|\chi^{(3)}|/2n_1^2$ . In the case of the synchronization  $n_1 \rightarrow n_0$ , and  $Q_0 \gg 1$ , one gets

$$\alpha_1^2(\omega_1) \approx 2n_2(n_1 - n_0)n_0^{-2}(1 - \omega_1/2\omega_0). \quad (15)$$

The angle distribution of the forward generated photons equals

$$N(\alpha_1) = e^{Q_0 - \alpha_1^4/\alpha_c^4}, \quad (16)$$

where  $\alpha_c = (\eta_0 n_2) / |n_2 - n_0| (2Q_0)^{1/2}$  is the characteristic angle. The spectrum of the photons is

$$N(\omega_1) \approx (n_2 / \omega_0 n_0) e^{Q_0 - (2\omega_0 - \omega_1)^2 / \sigma^2}, \quad \omega_1 \leq 2\omega_0, \quad (17)$$

where  $\sigma = \eta_0 \omega_0 n_0 / |(n_1 - n_0)(n_2 - n_0)| (2Q_0)^{1/2}$  is the characteristic width of the spectrum. Both,  $\alpha_c$  and  $\sigma$  are small:  $\sigma \ll \omega_0$ ,  $\alpha_c \ll 1$ . Consequently, in the case of synchronization the photon  $1\omega_1$  is emitted forward and the frequency is doubled. The total number of forward-generated photons equals  $N_{\text{tot}} = N_w \exp Q_0$ , where  $N_w \sim V \alpha_c \sigma / \lambda_1^3 \omega_0$  is the characteristic number of working modes  $\vec{k}_1$ ,  $\lambda_1 = c_1 / \omega_1$  is the linewidth,  $V$  is the excited volume. Practical threshold for the process corresponds to  $Q_0 \approx 30$ .

## 6. DISCUSSION

Frequency doubling was observed in such inverse-symmetrical media as glass fibres (see, e.g. [7-12]) and gases [13-15]. In both cases, to obtain a remarkable output, pre-illumination by a strong light was used; a very small output was obtained also without illumination [7, 8]. A conventional interpretation of the observed effects supposes the breaking of inverse symmetry by illumination. Although this interpretation seems to be commonly accepted nowadays, there are difficulties in explaining polarization data [10]: the polarization of the generated double-frequency wave in glass fibres follows the polarization of the initial wave and does not depend on the polarization of the preceding excitation. Noteworthy is here that the observed effect, both in glass fibres and gases, can be explained in the frames of the proposed theory without any symmetry-breaking assumption.

In gases, frequency doubling was observed under the excitation  $10^{14} - 10^{15} \text{ Wcm}^{-2}$  together with the multiphoton ionization (in xenon 11 photons are required for ionization) [13-15]. In [16] it was supposed that ionization builds up an electric field which breaks the inverse symmetry and creates a nonzero  $\chi^{(2)}$ . Note that the same ionization can strongly increase  $|\chi^{(3)}|$  without any symmetry breaking. Indeed, an estimation of  $|\chi^{(3)}|$  connected with the plasma  $\sim 5 \cdot 10^{15}$  elementary charges per  $\text{cm}^3$  created under experimental conditions [15], gives  $|\chi^{(3)}| \sim 10^{-21} \text{ cm}^2 \text{W}^{-1}$  (here  $|\chi^{(3)}|$  is resonantly enhanced due to small difference of  $\omega_2$  and plasma frequency  $\omega_p \approx 2 \cdot 10^{13} \text{ sec}^{-1}$ ). Taking into account that the duration of excitation pulses was  $t_0 = 30 \text{ ps}$ ,  $\omega_0 \approx 3 \cdot 10^{15} \text{ sec}^{-1}$ , and supposing that  $|1 - c_1/v_g| \sim 10^{-3}$ , one gets the threshold value  $Q_0 \approx 30$  for a peak power  $10^{14} \text{ Wcm}^{-2}$  in agreement with the experiment.

Typical parameters of the excitation used in the glass fibre experiments mentioned are: peak power  $\sim 30 \text{ kW}$ , mean frequency  $\omega_0 \sim 3 \cdot 10^{15} \text{ sec}^{-1}$ , pulse duration  $t_0 \sim 10^{-10} \text{ s}$ . The fibre diameter was  $\sim 10^{-3} \text{ cm}$ . Taking



$|\chi^{(3)}| \sim 10^{-17} \text{ cm}^2\text{W}^{-1} [1]$ ,  $n_0 \sim 1$ ,  $|1 - c_0/v_g| \sim 10^{-2}$ , one gets  $Q_0 \sim 7$ , which is rather close to the threshold values  $Q_0 \approx 30$  and which can explain the observed weak signal in nonilluminated fibres. To explain experimental data concerning the role of the preceding illumination, one should suppose that the illumination leads to an increase of  $|\chi^{(3)}|$  and does not lead to the breaking of symmetry. This explains also the polarization data mentioned.

The proposed interpretations of the observed effects mentioned as well as the presented estimations are, naturally, only preliminary and more thorough considerations with allowance made for actual experimental conditions are required.

In conclusion, we showed here that the use of the moving reference frame gives essential simplification of the description of quantum nonlinear optical processes caused by a strong plane wave excitation. By this method we obtained a nonperturbational description of the photon pairs generation. We showed that the angle distribution of forward-generated photons has a sharp peak in the synchronization limit; the frequency of these photons is close to the double frequency of the excitation. This process should be taken into account when explaining the frequency doubling observed in glass fibres and glasses.

## ACKNOWLEDGEMENT

The research was supported by the Estonian Science Foundation grant No. 369 and by an ISF grant.

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## **FOOTONPAARIDE GENEREERIMINE JA SAGEDUSE KAHEKORDISTUMINE LIKUVAS TAUSTSÜSTEEMIS**

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On esitatud teooria, mis kirjeldab erineva sagedusega footonite paaride genereerimist tugeva tasalaine poolt liikuvast taustsüsteemis. Niisuguses taustsüsteemis on tasalaine lainevektor null ja ergastav väli sõltub ainult ajast. See võimaldab kasutada ajast sõltuva keskkonna kvantkiirguse teooriat ja kirjeldada uuritavat protsessi ilma häiritusteooria abita. Erilist tähelepanu on pööratud niisugusele nelja laine segunemisele, kus kaks ühesugust footonit muunduvad kaheks erinevaks footoniks, millest üks on suunatud ettepoole ja teine tahapoole. See protsess võimaldab kahekordistada sagedust inversioonsümmeetrilises keskkonnas. Hinnangud näitavad, et kirjeldatud protsess võib seletada eksperimendis vaadeldud sageduse kahekordistumist klaaskiududes ja gaasides.

### I. INTRODUCTION

Advances in the synthesis of nanocrystals or clusters in solid media (see e.g. [1]) have opened interest in the study of size-dependent cluster properties, including optical characteristics. In particular, a cluster can possess unique optical properties depending on the size and shape of the cluster and the surrounding optical characteristics must be size-dependent. In [2], the impurity electronic levels in a one-dimensional cluster were calculated. In the present paper, a simple three-dimensional model of a cluster is considered to determine the size-dependent electronic levels of the impurity. The theory of impurity states in crystals developed the problem of impurity levels by the Green's function method [3]. The Green's function method is applied to the present cluster model. However, here the Green's function of a perturbed Hamiltonian is calculated whereas in [2] the Green's function of the unperturbed Hamiltonian was calculated. In [4], the impurity levels in a cluster were calculated by the method of W. M. Robertson [5].