### METHODICAL NOTES

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# Spatial configuration of light at consecutive nonlinear optical conversions

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<u>Abstract.</u> We present the results of analytical and computer predictions about the spatio-spectral configuration of the radiation generated at consecutive two-stage processes of parametric scattering and the sum frequency generation in a layered nonlinear crystal. A procedure for the derivation of analytical estimates, including the solution of the phase matching problem. Some quite bizarre spatio-frequency spectra are shown.

### 1. Introduction

Current advances in nonlinear quantum optics permit devising and realizing wonderful and sometimes exotic processes that could not even be dreamed of until recently. A prominent example from this area is brought to the readers' notice.

The emergence of nonlinear quantum optics is commonly associated with light frequency doubling [1]. The red beam of a ruby laser with the wavelength 694.2 nm was directed into a transparent quartz crystal, and an ultraviolet beam with the wavelength 347.1 nm emanated from the crystal. In this case, 'red' laser photon pairs transformed into single ultraviolet photons of the second harmonic. Approximately 15 years later it was determined that a peculiar ordering of the laser photon flux occurs in this case [2, 3]. Since pairs of photons are 'snatched out' of the laser beam, fluctuation intensity peaks are smoothed. In other words, the photon bunches are thinned. The shot noise in the detection of such a radiation can be suppressed. This is an amazing result, because it had

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Received 8 August 2005; revised 16 January 2006 Uspekhi Fizicheskikh Nauk **176** (5) 543–550 (2006) Translated by E N Ragozin; edited by A M Semikhatov been assumed for a rather long time that shot noise determined the limiting precision of measuring photoelectric devices and represented the so-called standard quantum limit. The nature of shot noise for a constant intensity of detectorilluminating light is related to the randomness of the detachment of a photoelectron from a photocathode, i.e., the photocathode has some minimal noise level (see, for instance, Ref. [4]).

The feasibility of light frequency doubling determines the feasibility of the inverse process as well [5]. This process was indeed realized in the so-called parametric light scattering [6], also in crystals, whereby single photons 'break down' to photon pairs. The pairs behave not only amazingly but also paradoxically. Being united by the common instant of production, the photons of each pair retain, like twins, exact interrelation even upon separation. For example, when one of the photons of a pair is recorded by a measuring device, the quantum state of the other changes instantaneously (!), even though the photons may be spaced at a huge distance: according to the universally accepted viewpoint, which has been borne out in experiment, the reduction occurs instantly (of course, to within the capability of the experimenter). The photons may fly several kilometers apart, but 'the information' about the results of detection of the first photon instantly changes the quantum state of the second one. Is it valid to say in this case that a supraluminal speed of information transmission is effected by way of parametric light scattering? Apparently not. The operation of a communication line between the remote observers of the photon pairs necessitates, apart from detectors, a 'telephone' as well: not knowing the results of the first photon detection, the observer of the second witnesses in effect a random signal (see, e.g., Ref. [7] and the references therein).

The light frequency doubling may be regarded as a special case of the summation of the frequencies of two beams:

$$\omega_1 + \omega_2 \to \omega_3 = \omega_1 + \omega_2 \,. \tag{1}$$

Equality (1) follows from the energy conservation law when two photons merge into one. Similarly, in the parametric



**Figure 1.** Diagram of consecutive parametric light conversion in a periodic structure consisting of domains with oppositely directed optical axes, which is formally equivalent to the change of sign in the quadratic nonlinearity coefficient  $\chi^{(2)}$ . The collimated laser pump beam with a circular radiation frequency  $\omega_2$  decays into beams with frequencies  $\omega'_1$  and  $\omega''_1$ , which satisfy relation (2). Each of these beams interacts with the laser beam to produce radiation with the sum frequency, i.e.,  $\omega'_3 = \omega_2 + \omega'_1$  and  $\omega''_3 = \omega_2 + \omega''_1$ .

scattering,

$$\omega_2 \to \omega_1' + \omega_1'' = \omega_2 \,. \tag{2}$$

Is it possible to consecutively realize both processes in a common crystal? In principle, yes, but it is quite difficult. The reason is that the velocities of all propagating beams should be equal, otherwise some beams would outrun the others and the nonlinear conversion efficiency would drastically decrease. But the dispersion effect should inhibit the fulfillment of the so-called phase matching condition. Furthermore, the crystal anisotropy has the effect that the phase velocities are different in different directions. Nevertheless, consecutive interaction is possible. This effect was investigated in crystals [8-10]. Arakelyan et al. [10] draw the conclusion, in particular, that the consecutive interaction may be highly efficient. However, employing periodic layered crystal structures provides additional degrees of freedom and considerable possibilities and at the same time complicates the theoretical solution to the problem [11] (Fig. 1). In this case, the spatial configuration of the radiation spectra takes on quite bizarre forms. Their description is the subject of this paper.

### 2. Parametric biphotons

We begin the formal description of consecutive light conversion with a parametric conversion where the frequencies of interacting light beams satisfy relation (2). In this case, the pump radiation photons with a frequency  $\omega_2$  decay into signal ( $\omega'_1$ ) and idler ( $\omega''_1$ ) photons, i.e., a down frequency conversion (from a higher frequency to lower ones) occurs.

We first assume that there are only three plane monochromatic radiation modes: the pump mode, which is characterized by the photon creation and annihilation operators  $\hat{a}_{2}^{\dagger}$  and  $\hat{a}_{2}$ , the signal mode  $(\hat{a}_{1}^{\prime\prime\dagger}, \hat{a}_{1}^{\prime\prime})$ , and the idler mode  $(\hat{a}_{1}^{\prime\prime\dagger}, \hat{a}_{1}^{\prime\prime})$ . Then, the operator  $\hat{a}_{2} \hat{a}_{1}^{\dagger} \hat{a}_{1}^{\prime\prime\dagger}$  takes the singlephoton pump mode  $|1\rangle_{2}$  and the vacuum  $|0\rangle_{1}$  of the signal and idler modes into a biphoton:  $\hat{a}_{2} \hat{a}_{1}^{\prime\dagger} \hat{a}_{1}^{\prime\prime\dagger} |1\rangle_{2} |0\rangle_{1}^{\prime} |0\rangle_{1}^{\prime\prime} = |0\rangle_{2} |1\rangle_{1}^{\prime} |1\rangle_{1}^{\prime\prime}$ . The Hermitian-conjugate operator  $\hat{a}_{2}^{\dagger} \hat{a}_{1}^{\prime} \hat{a}_{1}^{\prime\prime}$  performs the inverse transformation. These two mutually conjugate operators multiplied by constant coefficients make up the Hamiltonian of parametric interaction.

In a real situation, the interacting light beams are, of course, multimodal and have a certain spatial configuration and a finite spectral width. Therefore, light beams must be combined when they are resolved into plane monochromatic modes. The effective interaction Hamiltonian in a medium with a quadratic nonlinearity  $\chi^{(2)}$  can then be written as [13–16]

$$\begin{aligned} \hat{\mathcal{H}} &= -\frac{1}{2} \int_{V} \chi^{(2)}(\omega_{2}, \omega_{1}', \omega_{1}'', \mathbf{r}) \, \hat{E}_{2}^{(+)}(t, \mathbf{r}) \\ &\times \hat{E}_{1}^{\prime(-)}(t, \mathbf{r}) \, \hat{E}_{1}^{\prime\prime(-)}(t, \mathbf{r}) \, \mathrm{d}^{3}r + \mathrm{H.c.} \,, \end{aligned}$$
(3)

where the integration is performed over the crystal volume V, H.c. denotes the Hermitian-conjugate term, and the frequency-positive pump field operator

$$\hat{E}_{2}^{(+)}(t,\mathbf{r}) = \mathrm{i}\sum_{\mathbf{k}_{2}}\sqrt{\hbar\omega_{2}\left(\frac{2\pi}{L^{3}}\right)}\hat{a}_{\mathbf{k}_{2}}\exp\left[\mathrm{i}(\mathbf{k}_{2}\mathbf{r}-\omega_{2}t)\right] \quad (4)$$

is the sum of all possible plane modes in the quantization cube  $L^3$  (see, e.g., Ref. [17]), which are characterized by the wave vectors  $\mathbf{k}_2$  and the corresponding photon annihilation operators  $\hat{a}_{\mathbf{k}_2}$ .

Similarly, the frequency-negative operators of the signal and idler fields are given by

$$\hat{E}_{1}^{(-)}(t,\mathbf{r}) = -i\sum_{\mathbf{k}_{1}}\sqrt{\hbar\omega_{1}\left(\frac{2\pi}{L^{3}}\right)}\,\hat{a}_{\mathbf{k}_{1}}^{\dagger}\,\exp\left[-i(\mathbf{k}_{1}\mathbf{r}-\omega_{1}t)\right],$$
(5)

where the quantities with a subscript 1 should be marked with either a prime when the signal mode is described or a double prime in the description of the idler mode.

We now consider the consecutive interaction diagrammed schematically in Fig. 1. Because the frequencies  $\omega'_3$  and  $\omega''_3$  are higher than the pump frequency, this process is termed parametric amplification at low-frequency pumping, to distinguish it from parametric scattering, which is a down frequency conversion, when  $\omega'_1$  and  $\omega''_1$  are lower than  $\omega_2$ .

## **3. Parametric amplification at low-frequency pumping**

We thus consider the spatial effects in consecutive threefrequency processes. In this case, the interacting frequencies satisfy the relations

$$\begin{aligned}
\omega_2 &= \omega'_1 + \omega''_1, \\
\omega_2 &+ \omega'_1 &= \omega'_3, \\
\omega_2 &+ \omega''_1 &= \omega''_3.
\end{aligned}$$
(6)

The first process is the parametric amplification (or decay) at high-frequency pumping, whereby the beams with frequencies  $\omega'_1$  and  $\omega''_1$  are generated. In the next two processes, generation of the beams with the sum frequencies  $\omega'_3$  and  $\omega''_3$  occurs.

The first and the second processes in relations (6), as well as the first and the third, are consecutive, while the second and the third processes are simultaneous. The interaction Hamiltonian in the general form can then be written by analogy with the Hamiltonian that describes the production of two-photon light (3):

$$\begin{aligned} \hat{\mathcal{H}} &= -\frac{1}{2} \sum_{m,n} \int_{V} d^{3} \mathbf{r} \big[ \chi^{(2)}(\omega_{2}, \omega_{1m}', \omega_{1n}'', \mathbf{r}) \\ &\times \hat{E}_{2}^{(+)}(t, \mathbf{r}) \, \hat{E}_{1}^{\prime(-)}(t, \mathbf{r}) \, \hat{E}_{1}^{\prime(-)}(t, \mathbf{r}) \\ &+ \chi^{(2)}(\omega_{2}, \omega_{1m}', \omega_{3n}', \mathbf{r}) \, \hat{E}_{2}^{(+)}(t, \mathbf{r}) \, \hat{E}_{1}^{\prime(-)}(t, \mathbf{r}) \, \hat{E}_{3}^{\prime(-)}(t, \mathbf{r}) \\ &+ \chi^{(2)}(\omega_{2}, \omega_{1m}'', \omega_{3n}'', \mathbf{r}) \, \hat{E}_{2}^{(+)}(t, \mathbf{r}) \, \hat{E}_{1}^{\prime\prime(-)}(t, \mathbf{r}) \, \hat{E}_{3}^{\prime\prime(-)}(t, \mathbf{r}) \big] + \text{H.c.}, \end{aligned}$$
(7)

where

$$\hat{E}_{\alpha}^{(+)}(t,\mathbf{r}) = \mathrm{i}\sum_{\mathbf{k}_{\alpha}}\sqrt{\hbar\omega_{\alpha}\left(\frac{2\pi}{L^{3}}\right)}\,\hat{a}_{\alpha}\exp\left[\mathrm{i}(\mathbf{k}_{\alpha}\mathbf{r}-\omega_{\alpha}t)\right],\ (8a)$$

$$\hat{E}_{\alpha}^{(-)}(t,\mathbf{r}) = -i\sum_{\mathbf{k}_{\alpha}}\sqrt{\hbar\omega_{\alpha}\left(\frac{2\pi}{L^{3}}\right)}\,\hat{a}_{\alpha}^{\dagger}\exp\left[-i(\mathbf{k}_{\alpha}\mathbf{r}-\omega_{\alpha}t)\right].$$
(8b)

The first term in interaction Hamiltonian (7) corresponds to the first process in relations (6), and the second and the third terms correspond to the second and third processes in relations (6), respectively. In the approximation of collinear plane interacting modes, this process was studied in Refs [11, 12]. A spatially limited Gaussian pump for parametric interactions was considered in several papers (see, e.g., Refs [9, 16, 18–20]), although for monocrystalline nonlinear media only.

The pump is assumed to be fixed, classical, cylindrical, Gaussian, and monochromatic:

$$\hat{E}_2^{(-)} = E_2 \exp\left[-\left(\frac{r_\perp}{r_2}\right)^2 - \mathrm{i}(k_2 z - \omega_2 t)\right],\tag{9}$$

where  $E_2 = E_0 i \sqrt{\hbar \omega_2 (2\pi/L^3)}$  is the complex pump amplitude,  $r_2$  is the waist radius (in this case, the focal region is assumed to be substantially longer than the crystal length),  $\omega_2$ is the pump frequency, and  $k_2$  is its corresponding wavenumber.

We note that although the pump is monochromatic, the parametrically scattered beams 1' and 1" are broadband, with the effect that an additional double sum over m and n appears in Hamiltonian (7).

The solution of the Schrödinger equation

$$i\hbar \frac{d|\varphi\rangle}{dt} = \hat{H}|\varphi\rangle \tag{10}$$

can be formally written as

$$\left|\varphi(t)\right\rangle = \hat{U}|\varphi\rangle\,,\tag{11}$$

where  $|\varphi(t)\rangle$  is the field state vector. In the problem involved, the unitary evolution operator  $\hat{U}$  can be calculated using the perturbation theory technique. In the second order of the perturbation theory, it is given by (see, e.g., Ref. [20])

$$\hat{U} = \hat{I} + \frac{1}{\mathrm{i}\hbar} \int_{t_0}^{t} \mathrm{d}t' \hat{H}(t') + \left(\frac{1}{\mathrm{i}\hbar}\right)^2 \int_{t_0}^{t} \mathrm{d}t' \int_{t_0}^{t'} \mathrm{d}t'' \hat{H}(t') \hat{H}(t''),$$
(12)

where  $\hat{I}$  is the unit operator. From the normalization condition

$$\langle \varphi(t) | \varphi(t) \rangle \equiv \langle \varphi(t_0) | \varphi(t_0) \rangle,$$
 (13)

we have

$$\int_{t_0}^t \mathrm{d}t' \int_{t_0}^{t'} \mathrm{d}t'' \hat{H}(t') \, \hat{H}(t'') = \frac{1}{2} \left[ \int_{t_0}^t \mathrm{d}t' \hat{H}(t') \right]^2, \qquad (14)$$

where  $t_0$  and t are the instants of engagement and termination of the pump. As  $t_0 \rightarrow -\infty$  and  $t \rightarrow \infty$ , the integration with respect to t yields the delta function:

$$\int_{-\infty}^{\infty} dt \exp(i\omega t) = 2\pi\delta(\omega).$$
(15)

If the vacuum is at the input of the nonlinear crystal, it is transformed by the last term in expression (12) into the singlephoton states  $|1\rangle'_{3}$  and  $|1\rangle''_{3}$ . In the determination of the intensities of these states, of the  $\langle \varphi(t) | a_3^{\dagger} a_3 | \varphi(t) \rangle$  type, the nonlinearity is of the fourth order of smallness, although we restricted ourselves to only the second order in formula (12). Nevertheless, the accuracy of calculation by the perturbation theory is not exceeded because higher orders of expansion (12) (the third and the fourth) do not make contributions to the intensity of the field at the frequencies  $\omega_3$  in the fourth order in intensity. Indeed, the fourth order in intensity may appear, for instance, in a combination of the first and third orders in amplitude. But in the first order in amplitude, the state of the field at the frequencies  $\omega_2$  is the vacuum, and the intensity is zero. The same is true for the combination of the zeroth and the fourth orders in amplitude.

Integrating Hamiltonian (7) with respect to time yields the result

$$\begin{split} &\int_{-\infty}^{\infty} dt' \hat{H}(t') = \frac{2\pi^3}{L^3} \bigg\{ \sum_{m,n} \exp\left[ -\left(\mathbf{k}_{\perp 1m}' + \mathbf{k}_{\perp 1n}''\right)^2 \frac{r_2^2}{4} \right] \\ &\times F(\Delta k_1) \,\chi^{(2)}(\omega_2, \omega_{1m}', \omega_{1n}'') \sqrt{\omega_{1m}' \omega_{1n}''} \,\delta(\Delta \omega_1) \,\hat{a}_{1m}' + \hat{a}_{1n}'' + \\ &+ \sum_{m,n} \exp\left[ -\left(\mathbf{k}_{\perp 1m}' - \mathbf{k}_{\perp 3n}'\right)^2 \frac{r_2^2}{4} \right] \\ &\times F(\Delta k_2) \,\chi^{(2)}(\omega_2, \omega_{1m}', \omega_{3n}') \sqrt{\omega_{1m}' \omega_{3n}'} \,\delta(\Delta \omega_2) \,\hat{a}_{1m}' \,\hat{a}_{3n}'^{\dagger} \\ &+ \sum_{m,n} \exp\left[ -\left(\mathbf{k}_{\perp 1m}' - \mathbf{k}_{\perp 3n}'\right)^2 \frac{r_2^2}{4} \right] \\ &\times F(\Delta k_3) \,\chi^{(2)}(\omega_2, \omega_{1m}'', \omega_{3n}'') \sqrt{\omega_{1m}' \omega_{3n}''} \,\delta(\Delta \omega_3) \,\hat{a}_{1m}'' + \hat{a}_{3n}''^{\dagger} \bigg\} \\ &+ \text{H.c.} \,, \end{split}$$
(16)

where, for  $M = l/l_c$  periodic layers,

$$F(\Delta k) = \int_{0}^{l} dz \chi^{(2)}(z) \exp(iz\Delta k)$$
$$= \frac{\chi_{0}^{(2)}}{\Delta k} \left\{ 1 - \left[ -\exp(i\Delta k l_{c}) \right]^{M} \right\} \tan\left(\frac{\Delta k l_{c}}{2}\right), \quad (17)$$

*l* is the crystal length, and  $l_c$  is the layer length. Here, we have taken into account that the coupling coefficient  $\chi^{(2)}$  in the layered nonlinear crystal depends on *z*. In the integration, we used the relation

$$\int_{-\infty}^{\infty} d^2 \mathbf{r}_{\perp} \exp\left(-\frac{\mathbf{r}_{\perp}^2}{r_2^2} - \mathbf{i}\mathbf{k}_{\perp}\mathbf{r}_{\perp}\right) = \pi r_2^2 \exp\left(-\frac{k_{\perp}^2 r_2^2}{4}\right), (18)$$

where the infinite integration limits are justified when the transverse dimension of the nonlinear medium is far greater than the diameter of the pump waist.

The process peaks in efficiency when the quasi-phasematching condition

$$\Delta k l_{\rm c} = \pm \pi \,, \ \pm 3\pi \,, \dots \tag{19}$$

is satisfied, because  $F(\Delta k)$  in expression (17) is maximized in this case.

We can now write the expression for the vector of the quantum state at the frequencies  $\omega'_3$  and  $\omega''_3$  in the absence of a seed field at the frequencies  $\omega'_1, \omega''_1$  and  $\omega'_3, \omega''_3$ :

$$\begin{split} |\varphi_{3}\rangle &= \frac{1}{(i\hbar)^{2}} \int_{-\infty}^{\infty} dt' \hat{H}(t') \int_{-\infty}^{t'} dt \hat{H}(t) |0\rangle \\ &= \frac{1}{2(i\hbar)^{2}} \int_{-\infty}^{\infty} dt \hat{H}(t) \int_{-\infty}^{\infty} dt \hat{H}(t) |0\rangle \\ &= -\frac{2\pi^{6}}{L^{6}} r_{2}^{4} E_{2}^{2} \bigg\{ \sum_{m,n,p} \exp\left(-\left(\mathbf{k}_{\perp 1m}' + \mathbf{k}_{\perp 1n}''\right)^{2} \frac{r_{2}^{2}}{4}\right) \right. \\ &\times F(k_{2} - k_{z1m}' - k_{z1n}'') \\ &\times \chi^{(2)}(\omega_{2}, \omega_{1m}', \omega_{1n}'') \sqrt{\omega_{1m}' \omega_{1n}''} \,\delta(\omega_{2} - \omega_{1m}' - \omega_{1n}'') \\ &\times \bigg[ \exp\left(-\left(\mathbf{k}_{\perp 1m}' - \mathbf{k}_{\perp 3p}'\right)^{2} \frac{r_{2}^{2}}{4}\right) F(k_{2} + k_{z1m}' - k_{z3p}') \right. \\ &\times \chi^{(2)}(\omega_{2}, \omega_{1m}', \omega_{3p}') \sqrt{\omega_{1m}' \omega_{3p}''} \,\delta(\omega_{2} + \omega_{1m}' - \omega_{3p}') |1\rangle_{3p}' \\ &+ \exp\left(-\left(\mathbf{k}_{\perp 1m}' - \mathbf{k}_{\perp 3p}'^{2}\right)^{2} \frac{r_{2}^{2}}{4}\right) F(k_{2} + k_{z1m}' - k_{z3p}'') \\ &\times \chi^{(2)}(\omega_{2}, \omega_{1m}', \omega_{3p}'') \sqrt{\omega_{1m}' \omega_{3p}''} \\ &\times \chi^{(2)}(\omega_{2}, \omega_{1m}', \omega_{3p}'') \sqrt{\omega_{1m}' \omega_{3p}''} \bigg\} \,. \end{split}$$

From the standpoint of computer calculations, we emphasize that the choice of the quantization volume has a significant effect on the computation time because the step of calculation in k is equal to  $2\pi/L$ . The field should substantially decrease toward the boundary of the quantization volume, and therefore the length L of the quantization cube edge should exceed the diameter of the pump, at least in the transverse direction.

We replace the sums over *m* and *n* with integrals:

$$\begin{split} |\varphi_{3}\rangle &= -\frac{r_{2}^{4}E_{2}^{2}}{32} \\ \times \int_{-\infty}^{\infty} \mathbf{d}^{(3)}\mathbf{k}_{1}^{\prime} \int_{-\infty}^{\infty} \mathbf{d}^{(3)}\mathbf{k}_{1}^{\prime\prime} \exp\left(-\left(\mathbf{k}_{\perp 1}^{\prime} + \mathbf{k}_{\perp 1}^{\prime\prime}\right)^{2} \frac{r_{2}^{2}}{4}\right) \\ \times F\left(k_{2} - k_{z1}^{\prime} - k_{z1}^{\prime\prime}\right) \\ \times \chi^{(2)}(\omega_{2}, \omega_{1}^{\prime}, \omega_{1}^{\prime\prime}) \sqrt{\omega_{1}^{\prime}\omega_{1}^{\prime\prime}} \,\delta(\omega_{2} - \omega_{1}^{\prime} - \omega_{1}^{\prime\prime}) \\ \times \sum_{p} \left[\exp\left(-\left(\mathbf{k}_{\perp 1}^{\prime} - \mathbf{k}_{\perp 3p}^{\prime}\right)^{2} \frac{r_{2}^{2}}{4}\right) F\left(k_{2} + k_{z1}^{\prime} - k_{z3p}^{\prime}\right) \\ \times \chi^{(2)}(\omega_{2}, \omega_{1}^{\prime}, \omega_{3p}^{\prime}) \sqrt{\omega_{1}^{\prime}\omega_{3p}^{\prime}} \,\delta(\omega_{2} + \omega_{1}^{\prime} - \omega_{3p}^{\prime})|1\rangle_{3p}^{\prime} \\ + \exp\left(-\left(\mathbf{k}_{\perp 1}^{\prime\prime} - \mathbf{k}_{\perp 3p}^{\prime\prime}\right)^{2} \frac{r_{2}^{2}}{4}\right) F\left(k_{2} + k_{z1}^{\prime\prime} - k_{z3p}^{\prime\prime}\right) \\ \times \chi^{(2)}(\omega_{2}, \omega_{1}^{\prime\prime}, \omega_{3p}^{\prime\prime}) \sqrt{\omega_{1}^{\prime\prime}\omega_{3p}^{\prime\prime}} \,\delta(\omega_{2} + \omega_{1}^{\prime\prime} - \omega_{3p}^{\prime\prime})|1\rangle_{3p}^{\prime\prime} \right]. \end{split}$$

We express the primed  $k_z$  of the extraordinary waves in terms of the corresponding  $\omega$  for the optical crystal axis aligned with the y axis,

$$k_z = \sqrt{\left(\frac{n_{\rm e}\omega}{c}\right)^2 - k_x^2 - \left(\frac{n_{\rm e}}{n_{\rm o}} k_y\right)^2},\tag{22}$$

where *c* is the speed of light in empty space and the refractive indices of the extraordinary  $n_e$  and ordinary  $n_o$  waves are involved in Sellmeyer's formulas [21]

$$n_{\alpha}^{2} = a_{\alpha} + \frac{b_{\alpha}\omega^{2}}{(2\pi c)^{2} + c_{\alpha}\omega^{2}} - d_{\alpha}\left(\frac{2\pi c}{\omega}\right)^{2}, \qquad (23)$$

where  $a_{\alpha}$ ,  $b_{\alpha}$ ,  $c_{\alpha}$ , and  $d_{\alpha}$  are constants for the selected nonlinear crystal at a specific temperature and  $\alpha = 0$ , e. In the subsequent calculations, the coefficients  $a_{\alpha}$ ,  $b_{\alpha}$ ,  $c_{\alpha}$ , and  $d_{\alpha}$  are taken for the lithium niobate crystal [22].

We make a change of variables  $k_z \rightarrow \omega$  in expression (21) and integrate it with respect to  $\omega$ . Hence, we obtain the amplitude of the state  $|1\rangle'_3$  with the selected wave vector  $\mathbf{k}'_3$ :

$$\begin{aligned} \mathbf{A}_{3}^{\prime} &= -\frac{r_{2}^{*} E_{2}^{2}}{32} \,\chi^{(2)}(\omega_{2}, \omega_{3}^{\prime} - \omega_{2}, 2\omega_{2} - \omega_{3}^{\prime}) \\ &\times \chi^{(2)}(\omega_{2}, \omega_{3}^{\prime} - \omega_{2}, \omega_{3}^{\prime})(\omega_{3}^{\prime} - \omega_{2}) \sqrt{\omega_{3}^{\prime}(2\omega_{2} - \omega_{3}^{\prime})} \\ &\times \int_{-\infty}^{\infty} d^{2} \mathbf{k}_{\perp 1}^{\prime} \int_{-\infty}^{\infty} d^{2} \mathbf{k}_{\perp 1}^{\prime\prime} \\ &\times \left[ \exp \left\{ - \left[ (\mathbf{k}_{\perp 1}^{\prime} + \mathbf{k}_{\perp 1}^{\prime\prime})^{2} + (\mathbf{k}_{\perp 1}^{\prime} - \mathbf{k}_{\perp 3}^{\prime})^{2} \right] \frac{r_{2}^{2}}{4} \right\} \\ &\times \left| \frac{dk_{z1}^{\prime}}{d\omega_{1}^{\prime}} \right|_{\omega_{1}^{\prime} = \omega_{3}^{\prime} - \omega_{2}} \left| \frac{dk_{z1}^{\prime\prime}}{d\omega_{1}^{\prime\prime}} \right|_{\omega_{1}^{\prime\prime} = 2\omega_{2} - \omega_{3}^{\prime}} \\ &\times F \left( k_{2} - k_{z1}^{\prime} |_{\omega_{1}^{\prime} = \omega_{3}^{\prime} - \omega_{2}} - k_{z1}^{\prime\prime} |_{\omega_{1}^{\prime\prime} = 2\omega_{2} - \omega_{3}^{\prime}} \right) \\ &\times F \left( k_{2} + k_{z1}^{\prime} |_{\omega_{1}^{\prime} = \omega_{3}^{\prime} - \omega_{2}} - k_{z3}^{\prime\prime} |_{\omega_{3}^{\prime}} \right) \right]. \end{aligned}$$

$$(24)$$

The amplitude of the single-photon state mode (with two primes) is given by the same expression.

### 4. Analysis of the calculated relation for the amplitude of a state mode

To analytically estimate expression (24), it is expedient to make the following simplifying assumptions. From the totality of possible values of the wave vectors, we may select only those that correspond to the phase quasi-phase-matching conditions (19), i.e., satisfy the system of nonlinear equations

$$\sqrt{\left(\frac{(\omega_{3}-\omega_{2})n_{e}'}{c}\right)^{2}-k_{x3}^{2}-\left(\frac{n_{e}'}{n_{o}'}k_{y3}\right)^{2}} - \sqrt{\left(\frac{\omega_{3}n_{e}'''}{c}\right)^{2}-k_{x3}^{2}-\left(\frac{n_{e}''}{n_{o}'''}k_{y3}\right)^{2}} = -k_{2}\pm\frac{\pi+2\pi m}{l_{c}},$$
(25)
$$\sqrt{\left(\frac{(\omega_{3}-\omega_{2})n_{e}'}{c}\right)^{2}-k_{x3}^{2}-\left(\frac{n_{e}'}{n_{o}'}k_{y3}\right)^{2}} + \sqrt{\left(\frac{(2\omega_{2}-\omega_{3})n_{e}''}{c}\right)^{2}-k_{x3}^{2}-\left(\frac{n_{e}''}{n_{o}''}k_{y3}\right)^{2}} = k_{2}\pm\frac{\pi+2\pi n}{l_{c}},$$
(26)

and assume that the radiation at the frequency  $\omega_3$  occurs only for these values. Here,  $n'_{\alpha}$  is the refractive index at the frequency  $\omega_3 - \omega_2$ ,  $n''_{\alpha}$  is the refractive index at the frequency  $2\omega_2 - \omega_3$ , and  $n'''_{\alpha}$  is the refractive index at the frequency  $\omega_3$ . The integers *m* and *n* can be set equal to zero because the magnitude of *F* is, according to expression (17), inversely proportional to  $\Delta k$ , and therefore, even with a unit value of one of these integers, the intensity of the spectral component is lower by an order of magnitude than for their zero values. The system of equations (25), (26) is to be solved for  $k_{\perp 3} = \{k_{x3}, k_{y3}\}$  and  $\omega_3$ . The intensity of every such component of the spectrum is therefore determined by the equation

$$I(k_{\perp 3}, \omega_3) \sim \omega_3 (2\omega_2 - \omega_3)(\omega_3 - \omega_2)^2 \times \left[\chi^{(2)}(\omega_2, \omega_3 - \omega_2, 2\omega_2 - \omega_3)\chi^{(2)}(\omega_2, \omega_3 - \omega_2, \omega_3)\right]^2 \times \left|\frac{\mathrm{d}k_z}{\mathrm{d}\omega}\right|^2_{\omega=\omega_3-\omega_2} \left|\frac{\mathrm{d}k_z}{\mathrm{d}\omega}\right|^2_{\omega=2\omega_2-\omega_3}.$$
(27)

The solution of nonlinear system of equations (25), (26) in the general form is rather cumbersome; however, a compact analytical solution can be obtained in our special case.

We introduce coefficients A, B, C, D, E, and G as

$$A = \left(\frac{(\omega_3 - \omega_2) n'_{\rm e}}{c}\right)^2, \quad B = \left(\frac{(2\omega_2 - \omega_3) n''_{\rm e}}{c}\right)^2,$$
$$C = \left(\frac{\omega_3 n'''_{\rm e}}{c}\right)^2, \quad D = \left(\frac{n'_{\rm e}}{n'_{\rm o}}\right)^2,$$
$$E = \left(\frac{n''_{\rm e}}{n''_{\rm o}}\right)^2, \quad G = \left(\frac{n''_{\rm e}}{n''_{\rm o}}\right)^2$$
(28)

and introduce the notation for the right-hand sides of Eqns (25) and (26):

$$S = -k_2 \pm \frac{\pi + 2\pi m}{l_c}, \qquad T = k_2 \pm \frac{\pi + 2\pi n}{l_c}.$$
 (29)

System (25), (26) then becomes

$$\sqrt{A - x - Dy} - \sqrt{C - x - Gy} = S,$$
  
$$\sqrt{A - x - Dy} + \sqrt{B - x - Ey} = T,$$
 (30)

or

$$\sqrt{A - x - Dy} - S = \sqrt{C - x - Gy},$$
  
$$T - \sqrt{A - x - Dy} = \sqrt{B - x - Ey},$$
 (31)

where  $x = k_{x3}^2$  and  $y = k_{y3}^2$ . Because solutions are sought for real wavenumbers (*S* and *T* are real), the following inequalities must be satisfied:

$$\sqrt{A - x - Dy} \ge S,$$

$$C - x - Gy \ge 0,$$

$$\sqrt{A - x - Dy} \le T,$$

$$B - x - Ey \ge 0.$$
(32)

Hence, we obtain

$$A - Dy - 2S\sqrt{A - x - Dy} + S^{2} = C - Gy,$$
  

$$A - Dy - 2T\sqrt{A - x - Dy} + T^{2} = B - Ey.$$
(33)

With the notation  $Z = \sqrt{A - x - Dy}$  ( $Z \ge 0$ ), we eventually arrive at a system linear in Z and y:

$$A - Dy - 2SZ + S2 = C - Gy, A - Dy - 2TZ + T2 = B - Ey,$$
(34)

which has a solution

$$y = \frac{(A-B)/T - (A-C)/S + T - S}{(G-D)/S - (E-D)/T} .$$
 (35)

Next, passing to the variables x and y, it can be verified that the solution with both these quantities being positive and hence the quantities  $k'_{x3}$  and  $k'_{y3}$  simultaneously real exists only in several narrow frequency ranges.

Plotted in Fig. 2 are the functions  $k'_{x3}(\lambda_3)$  and  $k'_{y3}(\lambda_3)$  that are solutions of the system of equations (25), (26) for the laser radiation wavelength  $\lambda_2 = 1338$  nm. It is easily seen that the real solution of the system, determined by the intersection of the curves that represent these functions, exists only in several narrow wavelength domains  $\lambda_3$ . This signifies that the generation of light may be observed only in narrow spectral ranges and by no means everywhere. These are the values near 800, 940, and 1200 nm of all possible wavelengths  $\lambda_3$ , which reflects the fact that the radiation at a given wavelength has quite a specific directivity. Figure 3 shows the domain of solutions of system (25), (26) in the  $\{k'_{x3}, k'_{y3}, \lambda_3\}$  space for the second range, near the 940 nm wavelength. In a rather narrow wavelength range, from 942 to 943 nm, the  $k'_3$  projection values change from -0.4 to  $0.4 \ \mu m^{-1}$ , which gives values of the spatial angle about  $3.5^{\circ}$  with the z axis for radiation at this wavelength. A similar estimate for wavelengths in the vicinity of 800 nm yields a strictly forward direction, i.e., an angle of  $0^{\circ}$  with the z axis.

Therefore, in the first approximation, it is possible to estimate the radiation spectrum at the  $\omega_3$  frequency. But it is impossible to determine the spectral shape or its dependence on the number of layers of a periodically nonuniform medium in this way. To do this requires a more exact calculation of the



**Figure 2.** Transverse components of the wave vector  $\mathbf{k}'_3 = \{k'_{x3}, k'_{y3}\}$  (dashed and solid curves) that are real solutions of system of equations (25), (26). Radiation is produced at the wavelengths  $\lambda_3$  that correspond to intersections of the dashed and solid curves (domains I, II, and III).



**Figure 3.** The curve that corresponds to the solution of nonlinear system of equations (25), (26) in range II (see Fig. 2) for wavelengths  $\approx$  940 nm. The curve yields the interrelation between the direction of the generated light and its wavelength  $\lambda_3$ .

integral by formula (24). In this case, it is noteworthy that the domain of integration in which the integrand is nonzero is determined both by the exponential in the integrand and by the functions F.

The quantities

$$\frac{\mathrm{d}k'_{z1}}{\mathrm{d}\omega'_1}\Big|_{\omega'_1=\omega'_3-\omega_2} \text{ and } \left|\frac{\mathrm{d}k''_{z1}}{\mathrm{d}\omega''_1}\right|_{\omega''_1=2\omega_2-\omega'_3}$$

change only slightly in this domain and may therefore be factored out from the integrand. If the width of the Gaussian function

$$\exp\left\{-\left[(\mathbf{k}_{\perp 1}'+\mathbf{k}_{\perp 1}'')^2+(\mathbf{k}_{\perp 1}'-\mathbf{k}_{\perp 3}')^2\right]\frac{r_2^2}{4}\right\}$$

is substantially narrower than the width of the functions F, the exponential may be replaced with the delta function. We express the integrand in terms of the integration variables (in this case,  $k_{\perp 3}$  and  $\omega_3$  play the role of parameters) to obtain the field amplitude

$$\begin{aligned} A'_{3} &= -\frac{r_{2}^{4}E_{2}^{2}}{32} \chi^{(2)}(\omega_{2},\omega'_{3}-\omega_{2},2\omega_{2}-\omega'_{3}) \\ &\times \chi^{(2)}(\omega_{2},\omega'_{3}-\omega_{2},\omega'_{3})(\omega'_{3}-\omega_{2}) \\ &\times \sqrt{\omega'_{3}(2\omega_{2}-\omega'_{3})} \left| \frac{\mathrm{d}k'_{z1}}{\mathrm{d}\omega'_{1}} \right|_{\omega'_{1}=\omega'_{3}-\omega_{2}} \left| \frac{\mathrm{d}k''_{z1}}{\mathrm{d}\omega''_{1}} \right|_{\omega''_{1}=2\omega_{2}-\omega'_{3}} \\ &\times \int_{-\infty}^{\infty} \mathrm{d}k'_{x1} \, \mathrm{d}k''_{y1} \, \mathrm{d}k''_{y1} \, \mathrm{d}k''_{y1} \delta(k'_{x1}+k''_{x1}) \, \delta(k'_{y1}+k''_{y1}) \\ &\times \delta(k'_{x1}-k'_{x3}) \, \delta(k'_{y1}-k'_{y3}) \\ &\times F_{1}(k'_{x1},k'_{y1},k''_{x1},k''_{y1}) \, F_{2}(k'_{x1},k'_{y1},k''_{x1},k''_{y1}) \,, \end{aligned}$$
(36)

where

$$\begin{split} F_1 &= F\left(k_2 - k'_{z1}|_{\omega'_1 = \omega'_3 - \omega_2} - k''_{z1}|_{\omega''_1 = 2\omega_2 - \omega'_3}\right),\\ F_2 &= F\left(k_2 + k'_{z1}|_{\omega'_1 = \omega'_3 - \omega_2} - k'_{z3}|_{\omega'_3}\right). \end{split}$$

Introducing the function  $G = F_1F_2$ , we write the final expression

$$\begin{aligned} A'_{3} &= -\frac{r_{2}^{4} E_{2}^{2}}{32} \chi^{(2)}(\omega_{2}, \omega'_{3} - \omega_{2}, 2\omega_{2} - \omega'_{3}) \\ &\times \chi^{(2)}(\omega_{2}, \omega'_{3} - \omega_{2}, \omega'_{3})(\omega'_{3} - \omega_{2}) \\ &\times \sqrt{\omega'_{3}(2\omega_{2} - \omega'_{3})} \left| \frac{\mathrm{d}k'_{z1}}{\mathrm{d}\omega'_{1}} \right|_{\omega'_{1} = \omega'_{3} - \omega_{2}} \left| \frac{\mathrm{d}k''_{z1}}{\mathrm{d}\omega''_{1}} \right|_{\omega''_{1} = 2\omega_{2} - \omega'_{3}} \\ &\times G(k'_{x3}, k'_{y3}, -k'_{x3}, -k'_{y3}) \,. \end{aligned}$$
(37)

### 5. Spatial configuration of radiation spectra

The spatio – frequency spectrum of light at the frequency  $\omega$  is determined by the function *G* in formula (37). The spatial spectrum of radiation with the wavelength  $\lambda_3 = 805.2$  nm is



Figure 4. Intensity I of  $\lambda_3 = 805.2$  nm light as a function of its propagation direction for periodic layer numbers M = 10 (a) and M = 100 (b).



Figure 5. Intensity I of  $\lambda_3 = 942.5$  nm light as a function of its propagation direction for periodic layer numbers M = 10 (a) and M = 100 (b).

depicted in Fig. 4a for 10 layers. One can see that the radiation is primarily directed along the z axis and its divergence is equal to  $1-2^{\circ}$ , i.e., a bright spot is observable at the center surrounded by a halo of pale luminous rings.

Alternatively, at the light wavelength  $\lambda_3 = 942.5$  nm (Fig. 5), one can see one luminous ring with a sharper directivity,  $\approx 0.2^{\circ}$ . This result is to be compared with the result presented in Fig. 3, where the range of possible  $k_3$  values is confined to a relatively narrow interval.

It is also instructive to analyze how the increase in the number of crystal structure layers affects the spatial radiation spectrum (see Figs 4 and 5). The structure of the spectrum becomes more complicated: a large number of oscillations appear and the peaks become sharper. Therefore, with an increase in the number of layers, one might expect, on the one hand, a sharper picture and, on the other hand, an enrichment of the spectrum. In the former case (see Fig. 4), in particular the rings that fringe the central spot become brighter and, furthermore, both the rings and the spot itself are defined more clearly. In the latter case (see Fig. 5), the ring width becomes smaller, but the ring fringes, which were previously faintly visible, are more pronounced. This effect is attributable to the fact that a perfectly rigorous phase matching is, strictly speaking, possible only for an infinite number of



**Figure 6.** Numerically calculated spatial intensity distributions of the signal at the frequency  $\omega_3$  for different values of the pump radius *r*: curve l - r = 2 mm, 2 - r = 4 mm, 3 - r = 6 mm,  $4 - r = \infty$ .

layers. Decreasing the number of layers has the effect that the phase matching condition becomes less severe. Accordingly, the spatial configuration of the generated radiation and its directivity turn out to be smoother and more blurred.

The spatial pump configuration effect is similar. The larger its transverse dimensions, the sharper the radiation directivity. With decreasing the pump radius, the generated radiation 'spreads' in space. This follows explicitly from the resultant expression (24), in which the exponential involving the pump radius occurs in the integrand as a convolution with the function that describes the spatial radiation distribution for a plane pump. Figure 6 illustrates the character of how the spatial configuration of the generated radiation depends on the pump radius. The  $k_{x3}$ -projection value of the wave vector  $\mathbf{k}_3$  is plotted on the abscissa; in this case, the value of its projection on the y axis is taken to be zero ( $k_{y3} = 0$ ).

### 6. Conclusion

Despite the seemingly hopeless complexity of the problem, we have managed to solve it almost completely in the framework of an analytic approach. A computer was required only at the final stage to construct the spectra. This fortunate opportunity, which occurs rather infrequently in nonlinear optics and presented itself to us, impelled us to share these results with the reader. We hope that the results outlined above will be of interest not only to experts but also to a wide circle of people who take an interest in physics.

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#### References

- 1. Franken P A et al. Phys. Rev. Lett. 7 118 (1961)
- 2. Stoler D Phys. Rev. Lett. 33 1397 (1974)
- 3. Chmela P, Horák R, Peřina J J. Opt. Acta 28 1209 (1981)
- Akhmanov S A, D'yakov Yu E, Chirkin A S Vvedenie v Statisticheskuyu Radiofiziku i Optiku (Introduction to Statistical Radiophysics and Optics) (Moscow: Nauka, 1981)
- 5. Louisell W H Coupled Mode and Parametric Electronics (New York: Wiley, 1960)
- 6. Giordmaine J A, Miller R C Phys. Rev. Lett. 14 973 (1965)
- Belinskiĭ A V Usp. Fiz. Nauk 173 905 (2003) [Phys. Usp. 46 877 (2003)]
- 8. Tang C L Phys. Rev. 182 367 (1969)

- 9. Klyshko D N, Nazarova N I Zh. Eksp. Teor. Fiz. 58 878 (1970) [Sov. Phys. JETP 31 472 (1970)]
- 10. Arakelyan S M et al. Zh. Tekh. Fiz. 44 1253 (1974)
- 11. Chirkin A S et al. *Kvantovaya Elektron*. **30** 847 (2000) [*Quantum Electron*. **30** 847 (2000)]
- 12. Morozov E Yu, Chirkin A S J. Opt. A 5 233 (2003)
- 13. Klyshko D N Usp. Fiz. Nauk **154** 133 (1988) [Sov. Phys. Usp. **31** 74 (1988)]
- Belinskii A V, Klyshko D N Zh. Eksp. Teor. Fiz. 105 487 (1994) [JETP 78 259 (1994)]
- 15. Pittman T B et al. Phys. Rev. A 53 2804 (1996)
- 16. Belinskii A V Vestn. Mosk. Univ., Ser. 3. Fiz. Astron. (3) 34 (1999)
- Loudon R *The Quantum Theory of Light* (Oxford: Clarendon Press, 1973) [Tran slated into Russian (Moscow: Mir, 1976)]
- 18. Ito N, Inaba H *Opto-Electronics* **2** 81 (1970)
- Babin A A et al. Kvantovaya Elektron. 3 101 (1976) [Sov. J. Quantum Electron. 6 54 (1976)]
- Klyshko D N *Fotony i Nelineinaya Optika* (Photons and Nonlinear Optics) (Moscow: Nauka, 1980) [Translated into English (New York: Gordon and Breach, 1988)]
- 21. Nelson D F, Miculyak R M J. Appl. Phys. 45 3688 (1974)
- 22. Kitaeva G K et al. Appl. Phys. B 66 201 (1998)