

NONLINEAR-OPTICAL PHENOMENA

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Simultaneous nonlinear conversion of light in periodically poled crystals

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Abstract. **We consider a nondegenerate parametric process $\omega_3 \to \omega_1 + \omega_2$ and simultaneous sum frequency generation $\omega_1 + \omega_3 \to \omega_4$ and $\omega_2 + \omega_3 \to \omega_5$ in periodically poled quadratic crystals. An algorithm is proposed for the numerical description of the system evolution with full allowance for the quantum-mechanical interaction of all five plane monochromatic modes and pump depletion. The behaviour of the average number of photons of interacting modes and their mutual correlations is studied. An effect of inversion of the mutual correlation of the modes of the generated light is found.**

Keywords: *periodically poled crystals, nondegenerate parametric process, sum frequency generation.*

1. Introduction

One of the research directions of periodically poled crystals with a quadratic nonlinearity [1–7] is stipulated by the fact that the study of the quantum statistical characteristics of the light formed in such crystals is important not only for purely theoretical but also for applied problems of quantum information science. For example, the correlation characteristics of the generated light beams are used to produce quantum images [4, 5]. In solving such problems, the semi-classical field approximation (S method) is usually applied for the linearisation of nonlinear classical and operator equations [1, 2, 4, 5, 8]. This method makes it possible to obtain solutions in an analytical form, which in turn allows an analysis of interacting modes in thin periodically poled crystals with weak energy exchange. However, with increasing energy exchange and a noticeable depletion of the pump, the accuracy of the description is lost. The best approximation is given by perturbation theory (P method) [9, 10], but this is achieved by complicating the calculation by increasing the number of terms of higher orders. A more accurate quantum solution (Q method) can be obtained by numerical diagonalisation of the interaction Hamiltonian and finding the eigenvectors and eigenvalues of the quantum states [11–13]. It should also be noted that there is another method for solving problems of quantum nonlinear optics, which is based on polynomial algebra [14].

In the present paper, the quantum problem of the interaction of all plane monochromatic modes is solved with the depletion of the pump taken into account. The results differ from those obtained by approximate calculation methods in the case of strong energy exchange, when there is a noticeable depletion of the pump. The dynamics of the average number of photons in modes and their mutual correlation is shown, which is especially important for computer processing of information under noisy conditions, because the known correlation dependences allow one to extract efficiently a useful signal against the noise background, for example, when working with quantum ghost images [4, 5].

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2. Processes in a periodically poled crystal

Let five plane monochromatic modes characterised by the photon annihilation operators $\hat{a}_1$, $\hat{a}_2$, $\hat{a}_3$, $\hat{a}_4$ and $\hat{a}_5$ at optical frequencies ω_1, ω_2, ω_3, ω_4 and ω_5 collinearly propagate inside a periodically poled quadratic crystal. The operators satisfy the standard commutation relations: $[\hat{a}_j, \hat{a}_k^+] = \delta_{jk}$. At the same time, there occur three processes: parametric generation of subharmonics and frequency up-conversion by summing the frequencies of subharmonics with the pump frequency:

$$\omega_3 = \omega_1 + \omega_2, \qquad \delta k_3 = k_3 - k_1 - k_2 + m_3 G_3 = \Delta k_3 + m_3 G_3, \tag{1a}$$

$$\omega_1 + \omega_3 = \omega_4, \qquad \delta k_4 = k_4 - k_1 - k_3 + m_4 G_4 = \Delta k_4 + m_4 G_4, \tag{1b}$$

$$\omega_2 + \omega_3 = \omega_5, \qquad \delta k_5 = k_5 - k_2 - k_3 + m_5 G_5 = \Delta k_5 + m_5 G_5. \tag{1c}$$

Here, k_j are the moduli of the wave vectors of the mode with frequencies ω_j; $j = 1–5$; Δk_q is the wave detuning of the corresponding process for a homogeneous crystal; $q = 3, 4, 5$; $m_q = \pm 1, \pm 3, \pm 5, \ldots$ are the orders of quasi-phase matching; and $G_q = 2\pi/\Lambda_q$ is the wave number (modulus of the pseudovector) of the lattice of the domain structure with a period Λ_q.

The quasi-phase matching for processes (1a)–(1c) corresponds to $\Delta k_q = 0$. Simultaneous quasi-phase matching in the same domain structure with $G = G_3 = G_4 = G_5$ can be realised, for example, for different orders of quasi-phase matching m_q or for different coherence lengths $L_q = \pi/\Delta k_q = \Lambda_q/(2m_q)$. We numerically calculated the values of m_q and L_q for one domain structure with the wave number G when the equality $\delta k_q = 0$ is met to the fifth decimal place. The values of the quasi-phase-matching orders $m_{3,4} = 1$ [for processes (1a) and (1b)], $m_5 = 3$ [for process (1c) are found for coherence lengths $L_{3,4} \approx$

128.5 μm, $L_5 \approx 42.8$ μm and extraordinary wavelengths $\lambda_1 = 4.55$ μm, $\lambda_2 = 4.041$ μm, $\lambda_3 = 2.166$ μm, $\lambda_4 = 1.476$ μm, $\lambda_5 = 1.373$ μm in a periodically poled $LiNbO_3$ crystal.

Note that for the degenerate case the quasi-phase-matching condition was analysed in [6, 7], where the conditions for its realisation were found. We have calculated the quasi-phase matching for the case of nondegenerate generation.

The Hamiltonian of the interaction of the processes under consideration is represented in the form [1, 2]:

$$\hat{H}_{\text{int}} = h(\beta \hat{a}_1 \hat{a}_2 \hat{a}_3^+ + \gamma_1 \hat{a}_1 \hat{a}_3 \hat{a}_4^+ + \gamma_2 \hat{a}_2 \hat{a}_3 \hat{a}_5^+) + \text{H.c.}, \quad (2)$$

where h is the Planck constant; β and $\gamma_{1,2}$ are the coefficients of nonlinear interaction; and H.c. is the Hermitian conjugation. Expression (2) is an approximation of plane monochromatic modes under a collinear interaction. In this case, the transverse spatial structure of the beams is assumed to be homogeneous.

The operator equations of motion along the z axis inside the periodically poled crystal in the Heisenberg representation are described by the expression

$$\frac{\mathrm{d}\hat{a}_j}{\mathrm{d}z} = -\frac{\mathrm{i}}{h}[\hat{a}_j, \hat{H}_{\text{int}}]. \quad (3)$$

In the semi-classical field approximation, the pump is assumed to be classical with a constant amplitude $|B_3|$ and the phase φ_3: $B_3 = |B_3|\exp(\mathrm{i}\varphi_3)$.

We introduce the reduced interaction length $\zeta = \beta z$ and the dimensionless nonlinear coupling coefficients $\xi_{1,2} = \gamma_{1,2}/\beta$. The system of equations of motion becomes linear:

$$\frac{\mathrm{d}}{\mathrm{d}\zeta}\begin{pmatrix} \hat{a}_1 \\ \hat{a}_2^+ \\ \hat{a}_4 \\ \hat{a}_5^+ \end{pmatrix} = \begin{pmatrix} -\mathrm{i}B_3\hat{a}_2^+ - \mathrm{i}\xi_1 B_3^* \hat{a}_4 \\ \mathrm{i}B_3^*\hat{a}_1 + \mathrm{i}\xi_2 B_3 \hat{a}_5^+ \\ -\mathrm{i}\xi_1 B_3 \hat{a}_1 \\ \mathrm{i}\xi_2 B_3^* \hat{a}_2^+ \end{pmatrix}.$$

Its solutions can be found by means of the Laplace transform: $\hat{a}(\zeta) = Q(\zeta)\hat{a}(0)$, where $\hat{a}(\zeta) = (\hat{a}_1(\zeta), \hat{a}_2^+(\zeta), \hat{a}_4(\zeta), \hat{a}_5^+(\zeta))^{\mathrm{T}}$, T is the transpose operation, and $\hat{a}(0)$ is the vector of the mode operators at the input of the periodically poled crystal. The coefficients $Q(\zeta)$ differ somewhat from those given in [1, 3],

$$Q(\zeta) = \begin{pmatrix} \frac{C_2X_2 + C_1X_1}{2L_2} & \mathrm{i}\exp(\mathrm{i}\varphi_3)\frac{\sqrt{V_2}S_2 - S_1\sqrt{V_1}}{\sqrt{2}L_2} & \mathrm{i}\exp(-\mathrm{i}\varphi_3)\frac{S_1\sqrt{V_1}W_2 + S_2\sqrt{V_2}W_1}{2\sqrt{2}\xi_1 L_2} & \exp(\mathrm{i}2\varphi_3)\xi_2\frac{C_1 - C_2}{L_2} \\ -\mathrm{i}\exp(-\mathrm{i}\varphi_3)\frac{S_2\sqrt{V_2} - S_1\sqrt{V_1}}{\sqrt{2}L_2} & \frac{C_1W_1 + C_2W_2}{2L_2} & \exp(-\mathrm{i}2\varphi_3)\xi_1\frac{C_1 - C_2}{L_2} & -\mathrm{i}\exp(\mathrm{i}\varphi_3)\frac{\sqrt{V_2}S_2X_1 + S_1\sqrt{V_1}X_2}{2\sqrt{2}\xi_2 L_2} \\ -\mathrm{i}\xi_1\exp(\mathrm{i}\varphi_3)\frac{S_1\sqrt{V_2}X_1 + S_2\sqrt{V_1}X_2}{\sqrt{2}\sqrt{V_1V_2}L_2} & \exp(\mathrm{i}2\varphi_3)\xi_1\frac{C_2 - C_1}{L_2} & \frac{C_2W_1 + C_1W_2}{2L_2} & \mathrm{i}\sqrt{2}\xi_1\xi_2\exp(\mathrm{i}3\varphi_3)\frac{\sqrt{V_1}S_2 - \sqrt{V_2}S_1}{L_2\sqrt{V_1V_2}} \\ \exp(-\mathrm{i}2\varphi_3)\xi_2\frac{C_2 - C_1}{L_2} & -\mathrm{i}\exp(-\mathrm{i}\varphi_3)\frac{\sqrt{V_2}S_2X_1 + S_1\sqrt{V_1}X_2}{2\sqrt{2}\xi_2 L_2} & -\mathrm{i}\sqrt{2}\xi_1\xi_2\exp(-\mathrm{i}3\varphi_3)\frac{\sqrt{V_1}S_2 - \sqrt{V_2}S_1}{L_2\sqrt{V_1V_2}} & \frac{C_2X_1 + C_1X_2}{2L_2} \end{pmatrix}$$

since we have considered the more general case of arbitrary phases and amplitudes B_3. Here

$$L_1 = 1 - \xi_1^2 - \xi_2^2, \quad L_2 = \sqrt{\xi_1^4 + (-1 + \xi_2^2)^2 - 2\xi_1^2(1 + \xi_2^2)},$$

$$L_{3,4} = 1 \pm (\xi_1^2 \mp \xi_2^2),$$

$$C_{1,2} = \cosh\left(\frac{\zeta|B_3|\sqrt{V_{1,2}}}{\sqrt{2}}\right), \quad S_{1,2} = \sinh\left(\frac{\zeta|B_3|\sqrt{V_{1,2}}}{\sqrt{2}}\right),$$

$$V_{1,2} = L_1 \pm L_2, \quad W_{1,2} = L_2 \pm L_3, \quad X_{1,2} = L_2 \pm L_4.$$

In a more accurate approximation (in the perturbation theory approximation), the interaction of all five modes and the nonlinear terms of the interaction length are taken into account. The evolution operator in the Schrödinger representation for Hamiltonian (2) has the form

$$\hat{U}(\zeta) = \exp(-\mathrm{i}\hat{H}_{\text{int}}\zeta/h). \quad (4)$$

Because the Hamiltonian is in the exponent of the evolution operator, the Hamiltonian can be expanded in a Taylor series:

$$\hat{U}(\zeta) = \exp(-\mathrm{i}\hat{H}_{\text{int}}\zeta/h) = 1 + \left(-\mathrm{i}\frac{\hat{H}_{\text{int}}}{h}\zeta\right) + \frac{1}{2!}\left(-\mathrm{i}\frac{\hat{H}_{\text{int}}}{h}\zeta\right)^2 + \dots . \quad (5)$$

It was calculated up to the 12th order in the interaction parameter ζ, and the photon annihilation operators $\hat{a}_1$, $\hat{a}_2$, $\hat{a}_3$, $\hat{a}_4$ and $\hat{a}_5$ were found for five modes:

$$\hat{a}_{j\text{P}}(\zeta) = \hat{U}^+(\zeta)\hat{a}_j(0)\hat{U}(\zeta) = \hat{a}_j(0) + \left(\mathrm{i}\frac{\zeta}{h}\right)[\hat{H}_{\text{int}}, \hat{a}_j(0)] + \frac{1}{2!}\left(\mathrm{i}\frac{\zeta}{h}\right)^2[\hat{H}_{\text{int}}, [\hat{H}_{\text{int}}, \hat{a}_j(0)]] + \dots . \quad (6)$$

More precisely, the quantum problem can be solved as follows. We write down the matrix elements of the annihilation operator in the energy representation:

$$\hat{a}_{j_{n'n}} = \langle n'|\hat{a}_j|n\rangle = \delta_{n'n-1}\sqrt{n}. \quad (7)$$

Similarly, the matrix elements of the creation operator $\hat{a}_j^+$ are expressed. Then the interaction Hamiltonian (2) takes the form:

$$\hat{H}'_{\text{int}} = h(\beta \hat{a}'_1 \hat{a}'_2 \hat{a}'^+_3 + \gamma_1 \hat{a}'_1 \hat{a}'_3 \hat{a}'^+_4 + \gamma_2 \hat{a}'_2 \hat{a}'_3 \hat{a}'^+_5) + \text{H.c.}, \quad (8)$$

where

$$\hat{a}'_1 = (\hat{a}_{1_{n'_1 n_1}}) \otimes \hat{E}_2 \otimes \hat{E}_3 \otimes \hat{E}_4 \otimes \hat{E}_5;$$

$$\hat{a}'_2 = \hat{E}_1 \otimes (\hat{a}_{2_{n'_2 n_2}}) \otimes \hat{E}_3 \otimes \hat{E}_4 \otimes \hat{E}_5;$$

$$\hat{a}'_3 = \hat{E}_1 \otimes \hat{E}_2 \otimes (\hat{a}_{3_{n'_3 n_3}}) \otimes \hat{E}_4 \otimes \hat{E}_5;$$

$$\hat{a}'_4 = \hat{E}_1 \otimes \hat{E}_2 \otimes \hat{E}_3 \otimes (\hat{a}_{4_{n'_4 n_4}}) \otimes \hat{E}_5;$$

$$\hat{a}'_5 = \hat{E}_1 \otimes \hat{E}_2 \otimes \hat{E}_3 \otimes \hat{E}_4 \otimes (\hat{a}_{5_{n'_5 n_5}});$$

$\hat{E}_j$ are the identity matrices of the corresponding modes; $(\hat{a}_{j_{n'n}})$ is the matrix; and the sign $\otimes$ denotes the tensor product.

We diagonalise the interaction Hamiltonian (8) and find its eigenvectors and eigenvalues. The evolution operator in the matrix representation is calculated by the formula

$$\hat{U}(\zeta) = \sum_{m=0}^{M} \exp(-\mathrm{i}\lambda_m \zeta)|m\rangle\langle m|, \tag{9}$$

where $M = (n_1 + 1)(n_2 + 1)(n_3 + 1)(n_4 + 1)(n_5 + 1)$; n_j is the number of photons in modes; $|m\rangle$ is the eigenvector with the number m of photons for the eigenvalue λ_m of the interaction Hamiltonian (8): $\hat{H}'_{\text{int}}|m\rangle = \lambda_m|m\rangle$.

The density matrix is calculated from the formula

$$\hat{\rho}_{\text{Q}}(\zeta) = \hat{U}(\zeta)\hat{\rho}(0)\hat{U}^{+}(\zeta). \tag{10}$$

Here, $\hat{\rho}(0) = |\psi_0\rangle\langle\psi_0|$ is the density matrix of the initial state for all five modes at the input of the periodically poled crystal. In the tensor representation this matrix takes the form:

$$\hat{\rho}(0) = \hat{\rho}_1(0) \otimes \hat{\rho}_2(0) \otimes \hat{\rho}_3(0) \otimes \hat{\rho}_4(0) \otimes \hat{\rho}_5(0). \tag{11}$$

3. Mean values of the number of photons and correlation of modes

Let us calculate the average values of the number of photons and correlation coefficients of the second order, or the factor $g^{(2)}$, in the modes for the three methods in question by the formulas

$$N_{j(\text{S,P,Q})}(\zeta) = \begin{cases} \langle \hat{a}^{+}_{j(\text{S,P})}(\zeta)\,\hat{a}_{j(\text{S,P})}(\zeta)\rangle, \\ \langle \hat{a}'^{+}_{j(\text{Q})}(\zeta)\,\hat{a}'_{j(\text{Q})}(\zeta)\rangle, \end{cases} \tag{12}$$

$$g^{(2)}_{jk(\text{S,P,Q})}(\zeta) = \begin{cases} \dfrac{\langle \hat{a}^{+}_{j(\text{S,P})}(\zeta)\,\hat{a}_{j(\text{S,P})}(\zeta)\,\hat{a}^{+}_{k(\text{S,P})}(\zeta)\,\hat{a}_{k(\text{S,P})}(\zeta)\rangle}{N_{j(\text{S,P})}(\zeta)\,N_{k(\text{S,P})}(\zeta)}, \\ \dfrac{\langle \hat{a}'^{+}_{j(\text{Q})}(\zeta)\,\hat{a}'_{j(\text{Q})}(\zeta)\,\hat{a}'^{+}_{k(\text{Q})}(\zeta)\,\hat{a}'_{k(\text{Q})}(\zeta)\rangle}{N_{j(\text{Q})}(\zeta)\,N_{k(\text{Q})}(\zeta)}. \end{cases} \tag{13}$$

Here, the subscripts for $g^{(2)}$ denote the mode numbers for which the correlation coefficient is calculated, and the calculation method (S, P, Q).

The calculations were performed for $\xi_1 = 0.6$ and $\xi_2 = 0.4$, when the modes were in the state $|\psi_0\rangle = |n_{10}\rangle|n_{20}\rangle|\alpha_{30}\rangle|n_{40}\rangle|n_{50}\rangle$ at the input ($\zeta = 0$) of the periodically poled crystal. It was assumed that modes 1, 2, 4, 5 were in the vacuum state $|0\rangle$, and the pump was in the coherent state with an average number of photons $|\alpha_{30}|^2 = 3$ and phase $\varphi_{30} = \pi/3$. The constant initial phase of the pump affects the calculation results, because it determines the conditions for the entry of radiation into a nonlinear crystal.

For comparison, we also carried out a calculation for all modes, except pump modes, in single-photon states. In this case, the general trends described below are preserved.

The correctness of the computations was verified by controlling the commutation relations

$$[\hat{a}_j(\zeta), \hat{a}^{+}_j(\zeta)] = 1 \quad \text{и} \quad [\hat{a}'_j(\zeta), \hat{a}'^{+}_j(\zeta)] = 1.$$

4. Results and conclusions

Figures 1 and 2 show the dynamics of the average number of photons and correlation coefficients, or, in other words, the factor $g^{(2)}$, inside the periodically poled crystal for different calculation methods. It can be seen that the S method, like the P method, appreciably loses its accuracy when the pump is depleted compared to the more accurate quantum calculation method (Q).

The curves in Fig. 1 show almost periodic oscillations of the average number of photons, in contrast to the curves obtained by the approximate methods S and P. At the initial stage of the interaction, we first observe the nondegenerate parametric process $\omega_3 \to \omega_1 + \omega_2$ of the decay of the photon of mode 3 into two photons of modes 1 and 2, and then the sum frequency generation $\omega_1 + \omega_3 \to \omega_4$ and $\omega_2 + \omega_3 \to \omega_5$. Then, the reverse processes occur, and they compete with each other, which is clearly seen from the oscillating character of the curves for $N_{1\text{Q}}$, $N_{2\text{Q}}$, $N_{3\text{Q}}$, $N_{4\text{Q}}$, $N_{5\text{Q}}$ under strong energy exchange, i.e., with increasing ζ.

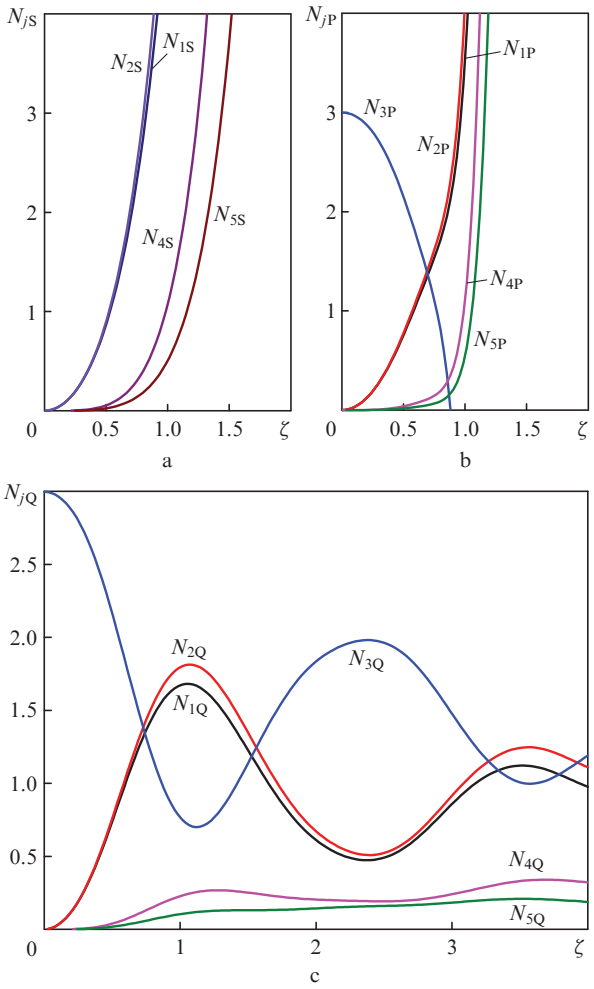

Figure 1. Dynamics of the average number of photons in modes, calculated by (a) S, (b) P and (c) Q methods.

For comparison, we have also calculated the average mode intensity in the classical description, taking into account the depletion of the pump, and also obtained appreciable discrepancies with the quantum computation presented here.

Figure 2 shows the values of the correlation coefficients, or second-order correlation factors. It can be seen that the S and P methods lose accuracy even at a relatively small depletion of the pump. The curves calculated by the more accurate Q method behave almost like periodic functions. At $g^{(2)} > 1$, pair correlated photons in two modes predominate, whereas

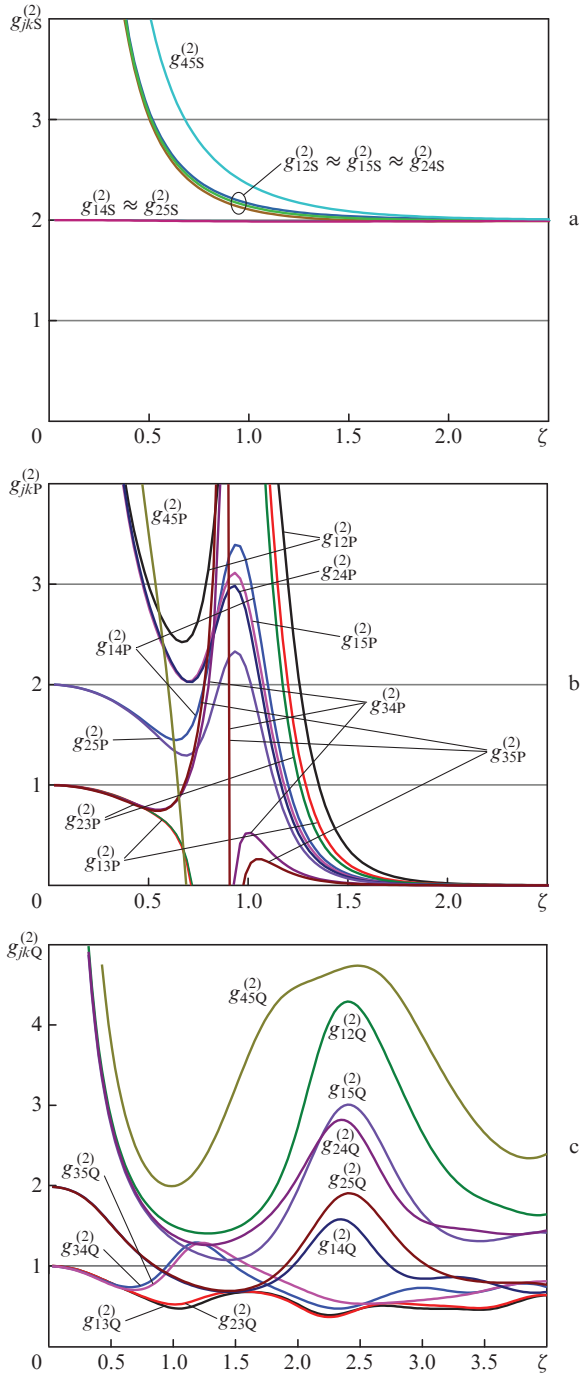

Figure 2. (Colour online) Correlation coefficients calculated by (a) S, (b) P and (c) Q methods.

at $g^{(2)} < 1$, single uncorrelated photons predominate in the same way as the single mode $g^{(2)} > 1$ corresponds to the grouping and super-Poisson statistics of photons, and $g^{(2)} < 1$ corresponds to anti-grouping and sub-Poisson statistics [15, 16].

One can see from Fig. 2, as well as from Fig. 1 that process (1a) is first realised, followed by processes (1b) and (1b). With strong energy exchange, all three processes begin to compete with each other, which manifest themselves in an almost periodic nature of the given curves. As a result of process (1b), it is practically impossible to simultaneously observe pair coincidences of photon modes 1 and 3, and 2 and 3, which confirms the conclusions of [4]. On the other hand, pair modes 1 and 2, 1 and 5, 2 and 4, and 4 and 5 are significantly correlated and can be used to reconstruct images using computer processing of information by algorithms proposed in [4, 5]. It is very important to note that between modes 1 and 4, 2 and 5, 3 and 4, and 3 and 5 there can be both a mutual correlation and an anticorrelation of photons, which is manifested at $g^{(2)} < 1$. To realise this or other mode, it is necessary to choose the appropriate interaction length, taking into account an intense energy exchange between the modes. This effect of inversion of mutual correlation was established by us on the basis of the described quantum calculation of the interaction of all five modes, and not only in the case of a vacuum state at the input of all modes except pumping, but also in the case of single-photon seeding. As follows from Figs 2a and 2b, it is practically impossible to detect it by known approximate methods.

Thus, we have solved the problem of quantum description of the interaction of plane monochromatic modes in a periodically poled crystal. For comparison, the same problem has been solved by the method of perturbation theory and in the semi-classical field approximation. The similarity of the results for all three methods has been observed only at the initial stage of evolution when the depletion of the pump is insignificant. Considerable discrepancies in question have been shown to appear at a later stage. The results of quantum calculations for intense energy exchange between plane monochromatic modes have been obtained by the numerical method taking into account the depletion of the pump.

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