
 ATOMS, MOLECULES, OPTICS

SPONTANEOUS PARAMETRIC DOWN-CONVERSION IN BIAXIAL CRYSTALS: PECULIARITIES OF THE POLARIZATION STATE

© 2024 D. N. Frolovtssev*, S. A. Magnitsky

Lomonosov Moscow State University, Faculty of Physics
Moscow, 119991, Russia

*e-mail: dfrolovtssev@gmail.com

Received September 06, 2023

Revised September 23, 2023

Accepted September 25, 2023

Abstract. A consistent analysis of the quantum state of polarization of SPDC radiation is presented and the peculiarities of the quantum state of polarization of SPDC in biaxial nonlinear optical crystals are considered. It is shown that the SPDC polarization deviation angle can exceed 15° , and the angle between the signal and idler wave vectors \mathbf{D} can exceed 30° . Estimates of the curvature of the cone formed by SPDC radiation in biaxial crystals are also given. The influence of SPDC polarization deviation in a non-collinear mode on the entanglement of bi-photon states generated by a double-crystal scheme is analyzed, it is shown that Tangle of the generated quantum state can deteriorate by 6%, and conditions are identified under which entanglement can be completely restored.

Keywords: Spontaneous parametric down-conversion, polarization deviation, biaxial crystals, double-crystal scheme

DOI: 10.31857/S00444510240104e8

1. INTRODUCTION

Spontaneous parametric down-conversion [1] (SPDC) is the effect of the generation of a pair of photons as a result of three-wave interaction in a quadratic nonlinear medium of the pump wave and fluctuations of the electromagnetic vacuum. At the same time, the laws of conservation of energy and momentum are fulfilled, which can be written as:

$$\begin{aligned}\hbar\omega_p &= \hbar\omega_s + \hbar\omega_i \\ \hbar\mathbf{k}_p &= \hbar\mathbf{k}_s + \hbar\mathbf{k}_i,\end{aligned}\quad (1)$$

where $\omega_{p,s,i}$ and $\mathbf{k}_{p,s,i}$ are the frequency and wave vector of pump waves (p), signal (s) and idle (i) waves respectively.

In the case of the first type of SPDC, which is considered in this paper, pumping is a wave of the same type, and the signal and idle waves are of a different type. In uniaxial crystals, the signal and optical waves are either *ordinary* or *extraordinary*, and in biaxial crystals they are either *fast* or *slow*.

The SPDC effect occupies one of the central places in modern quantum optical technologies and research [2, 3]. Thus, the SPDC effect is used in metrology for the non-etalon determination of the quantum efficiency [4] of single photon detectors [5–7], based on the SPDC, methods for measuring distances with accuracy above the standard quantum limit are being developed [8]. SPDC

occupies a special place in quantum technologies [9]. The SPDC sources of photon pairs entangled by polarization are the bricks for the realization of multiphoton entangled quantum states. This method was used for the first time to obtain the Greenberger–Horn–Zeilinger (HHZ) state [10] of three polarization-entangled photons, and later it was possible to obtain the maximally entangled quantum state of 12 photons, each of which was in a separate spatial mode [11].

One of the key schemes for generating polarization-entangled photon pairs is a two-crystal scheme [12] using SPDC with the first type of synchronism in a non-collisional mode. The non-collinear mode has an advantage over the collinear mode (for example, when used in crystals with a regular domain structure [13,14]) in that it allows controlling the frequency [15–17] and angular [18, 19] degrees of freedom of the quantum state due to a change in the scattering angle [20]. Quantum states with a high degree of quantum entanglement have also been obtained using the non-collinear mode [21–23]. However, in the non-collinear mode, the question about the direction of polarization of the wave, unlike the collinear mode, becomes non-trivial.

In the non-collinear geometry of the SPDC, the wave vectors \mathbf{k}_s and \mathbf{k}_i are not parallel to \mathbf{k}_p , and the direction of oscillations of the vectors \mathbf{E} and \mathbf{D} of the signal and idle

waves depend on the direction of scattering, which is the phenomenon of deviation of the polarization of the SPDC. If for “ordinary” nonlinear optics the phenomenon of polarization deviation does not lead to qualitative changes in the process of parametric generation, then in the case of SPDC it becomes significant when trying to build two-crystal circuits with a high degree of entanglement of the quantum polarization state [24,25]. The direction of oscillations of vectors \mathbf{E} and \mathbf{D} in uniaxial crystals has been studied in the literature [26], however, for biaxial crystals, as far as we know, the question of polarization SPDC deviation has not been studied. The main attention was paid to the numerical solution of the Fresnel equation and the determination of the refractive index in biaxial crystals to calculate the synchronism of second harmonic generation [27-29] and SPDC [30]. Note that the interest in biaxial crystals is determined by the fact that in some biaxial crystals, the value of effective nonlinearity exceeds the value of effective nonlinearity in uniaxial crystals. For example, in a BiBO crystal, the value of effective nonlinearity is twice higher (≈ 3.5 pm/V) than in BBO (≈ 1.75 pm/V) [31] for a frequency-degenerate SPDC with pumping at a wavelength of 405 nm. The direction of the polarization plane of the SPDC radiation plays a special role in the new field of phantom polarimetry [32], which uses SPDC sources operating in a non-collinear mode [33,34]. Deviation of the radiation polarization plane in the object arm in the phantom polarimeter will lead to a systematic error in the determination of the azimuth angle of the anisotropy of the object under study.

The purpose of this work is to obtain an expression for the deviation of the direction of oscillations of the vector \mathbf{D} of the SPDC radiation in biaxial crystals and to determine the degree of influence of polarization deviation on the entanglement of quantum states generated by a two-crystal circuit using biaxial crystals.

In section 2, the expressions for the direction of oscillations of the vectors \mathbf{E} and \mathbf{D} of natural waves in birefringent crystals are given. We proceeded from the fact that the direction of oscillation of the vector \mathbf{D} of its own wave is the semi-axis of the ellipse section of an ellipsoid of wave normal with a plane perpendicular to the vector \mathbf{k} . The solution includes both the case of uniaxial and the case of biaxial crystals. In section 3, the expressions for the quantum polarization state of the SPDC radiation in collinear and non-collinear modes are given.

In section 4, numerical estimates of the effect of the polarization deviation of the SPDC in a two-crystal scheme for

a biaxial BiBO crystal are given and a comparison is made for the case of a uniaxial BBO crystal. It is shown that in a two-crystal scheme using BiBO crystals, the coupling parameter is due to the phenomenon of polarization deviation is deteriorating, and conditions have been identified under which the deterioration of cohesion can be fully restored. In section 5, the results of the work are summarized.

2. POLARIZATION OF LIGHT IN BIAxIAL CRYSTALS

To find the direction of oscillations of the vector \mathbf{D} and determine the value of the refractive index, we will use the equation of the ellipsoid of wave normals [35-37]

$$\frac{X^2}{\varepsilon_x} + \frac{Y^2}{\varepsilon_y} + \frac{Z^2}{\varepsilon_z} = 1, \quad (2)$$

where $\varepsilon_{x,y,z}$ — are the main components of the dielectric constant tensor, $\{X,Y,Z\}$ — is a crystal-physical coordinate system in which the dielectric permeability tensor has a diagonal appearance. To find the direction of oscillation of vector \mathbf{D} is a crystal-physical coordinate system in which the dielectric permeability tensor has a diagonal appearance. To find the direction of oscillation of vector $\mathbf{k} = \{k_X, k_Y, k_Z\}$. The cross section is an ellipse, the main semi-axes of which set the direction of vibrations \mathbf{D} , and their lengths are equal to the values of the refractive indices of the corresponding waves.

The equation of the secant plane has the form

$$(\mathbf{k}, \mathbf{r}) = Xk_X + Yk_Y + Zk_Z = 0. \quad (3)$$

We carry out the procedure for finding the direction of rotation of the vector \mathbf{D} and the refractive indices using the affine transformation of the system of coordinates [38], in which the ellipsoid of wave normal lines will have the form of a sphere of unit radius. To do this, we will make the following substitution of variables $(X,Y,Z) \rightarrow (u,v,w)$:

$$u = X / \sqrt{\varepsilon_x}, \quad v = Y / \sqrt{\varepsilon_y}, \quad w = Z / \sqrt{\varepsilon_z}, \quad (4)$$

$$u^2 + v^2 + w^2 = 1.$$

In this case, the equation of the secant plane in the new coordinate system will have the form

$$k_X \sqrt{\varepsilon_x} u + k_Y \sqrt{\varepsilon_y} v + k_Z \sqrt{\varepsilon_z} w = 0. \quad (5)$$

The vector perpendicular to the secant plane, in the new coordinate system can be written as

$$\kappa = (k_X \sqrt{\epsilon_x}, k_Y \sqrt{\epsilon_y}, k_Z \sqrt{\epsilon_z}) = (\kappa_u, \kappa_v, \kappa_w). \quad (6)$$

In the case of $k_X^2 + k_Y^2 = 0$, $k_Z \neq 0$ in an anisotropic medium, two eigenwaves with directions of oscillation of the vector \mathbf{D} can propagate along the axes X and Y and with refractive indices of $\sqrt{\epsilon_x}$ and $\sqrt{\epsilon_y}$ respectively.

Considering the case of $k_X^2 + k_Y^2 \neq 0$. Let's find the equation of the secant figure in parametric form. For the case of the coordinate system $\{u, v, w\}$ the secant figure will be a circle of unit radius lying in a plane perpendicular to κ . It is not difficult to check by direct substitution that the two following vectors are perpendicular to k and each other:

$$\begin{aligned} e_1 &= \frac{1}{\sqrt{\kappa_u^2 + \kappa_v^2}} \begin{pmatrix} \kappa_v \\ -\kappa_u \\ 0 \end{pmatrix}, \\ e_2 &= \frac{[\kappa \times e_1]}{|\kappa|} = \frac{1}{\sqrt{(\kappa_u^2 + \kappa_v^2)(\kappa_u^2 + \kappa_v^2 + \kappa_w^2)}} \times \\ &\quad \times \begin{pmatrix} \kappa_u \kappa_w \\ \kappa_v \kappa_w \\ -(\kappa_u^2 + \kappa_v^2) \end{pmatrix}. \end{aligned} \quad (7)$$

Thus, in the coordinate system $\{u, v, w\}$ the equation of the secant figure in parametric form is written as

$$\mathbf{r}_{\{u,v,w\}} = \mathbf{e}_1 \sin s + \mathbf{e}_2 \cos s, \quad (8)$$

where s is the parameter, taking values from 0 to 2π . In the original notation

$$\begin{aligned} \mathbf{e}_1 &= \frac{1}{\sqrt{k_X^2 \epsilon_x + k_Y^2 \epsilon_y}} \begin{pmatrix} k_Y \sqrt{\epsilon_y} \\ -k_X \sqrt{\epsilon_x} \\ 0 \end{pmatrix}, \\ \mathbf{e}_2 &= \frac{1}{\sqrt{(k_X^2 \epsilon_x + k_Y^2 \epsilon_y)(k_X^2 \epsilon_x + k_Y^2 \epsilon_y + k_Z^2 \epsilon_z)}} \times \\ &\quad \times \begin{pmatrix} k_X k_Z \sqrt{\epsilon_x \epsilon_z} \\ k_Y k_Z \sqrt{\epsilon_y \epsilon_z} \\ -(\kappa_X^2 \epsilon_x + \kappa_Y^2 \epsilon_y) \end{pmatrix}. \end{aligned} \quad (9)$$

In the original notation $\{u, v, w\} \rightarrow \{X, Y, Z\}$, $\mathbf{e}_1 \rightarrow \mathbf{f}_1$, $\mathbf{e}_2 \rightarrow \mathbf{f}_2$ we have

$$\begin{aligned} \mathbf{f}_1 &= \sqrt{\frac{\epsilon_x \epsilon_y}{k_X^2 \epsilon_x + k_Y^2 \epsilon_y}} \begin{pmatrix} k_Y \\ -k_X \\ 0 \end{pmatrix}, \\ \mathbf{f}_2 &= \sqrt{\frac{\epsilon_z}{(k_X^2 \epsilon_x + k_Y^2 \epsilon_y)(k_X^2 \epsilon_x + k_Y^2 \epsilon_y + k_Z^2 \epsilon_z)}} \times \\ &\quad \times \begin{pmatrix} k_X k_Z \epsilon_x \\ k_Y k_Z \epsilon_y \\ -(\kappa_X^2 \epsilon_x + \kappa_Y^2 \epsilon_y) \end{pmatrix}. \end{aligned} \quad (10)$$

In this case, the equation of the ellipse obtained as a result of the section of the Fresnel ellipsoid with a plane perpendicular to \mathbf{k} in the coordinate system $\{X, Y, Z\}$ has the form

$$\mathbf{r}(X, Y, Z) = \mathbf{f}_1 \cos s + \mathbf{f}_2 \sin s, \quad (11)$$

where s is a parameter taking values from 0 to 2π . The directions of its main semi-axes determine the direction of oscillations of the vector \mathbf{D} , and the lengths of the semi-axes are equal to the corresponding refractive indices.

In uniaxial crystals, $\epsilon_x = \epsilon_y$, and $(\mathbf{f}_1, \mathbf{f}_2) = 0$, i.e. the vectors \mathbf{f}_1 and \mathbf{f}_2 are the semi-axes of the ellipse and set the directions of oscillation of the vector \mathbf{D} , and their lengths determine the refractive indices. It is also noted that the vector \mathbf{f}_1 , which determines the direction of oscillations of the vector \mathbf{D} and the refractive index for an ordinary wave, lies in the (XY) plane. At the same time, its length does not depend on the direction \mathbf{k} , which coincides with the known result for showing the refraction of an ordinary wave in birefringent crystals.

However, in biaxial crystals, in which all the three main components of the dielectric constant tensor are not equal to each other, $(\mathbf{f}_1, \mathbf{f}_2) \neq 0$, and the main semi-axes of the ellipse are determined by the expression (11), $\mathbf{r}(s_{1,2,3,4})$ at parameter values

$$\begin{aligned} s_1 &= \frac{\pi}{4} - \frac{1}{2} \arctg \frac{|\mathbf{f}_1|^2 - |\mathbf{f}_2|^2}{(\mathbf{f}_1, \mathbf{f}_2)}, \\ s_2 &= s_1 + \pi / 2, \\ s_{3,4} &= t_{1,2} + \pi, \end{aligned} \quad (12)$$

in this case, $\mathbf{r}(s_{3,4})$ and $\mathbf{r}(s_{1,2})$ are respectively equal in modulus and oppositely directed.

It is noteworthy that in the coordinate system $\{X, Y, Z\}$ the tensor ϵ_{ij} is diagonalized and the inverse tensor to it ϵ_{ij}^{-1} has the form

$$\epsilon_{ij}^{-1} = \begin{pmatrix} \epsilon_x^{-1} & 0 & 0 \\ 0 & \epsilon_y^{-1} & 0 \\ 0 & 0 & \epsilon_z^{-1} \end{pmatrix}. \quad (13)$$

Considering that

$$\mathbf{E} = \frac{\epsilon_{ij}^{-1}}{\epsilon_0} \mathbf{D},$$

having the coordinates of the vector \mathbf{D} , it is not difficult to calculate the coordinates of the vector \mathbf{E} .

3. THE STATE OF SPDC RADIATION POLARIZATION

3.1. Collinear scattering mode

With synchronization of the first type, when the frequencies of the signal and idle waves are equal, the collinear scattering mode corresponds to the so-called *degenerate* mode, i.e. as a result of the SPDC, two photons are generated in the same mode [39]. The quantum polarization state of the SPDC radiation has the form

$$|\psi\rangle = \mathbf{p}_{k_s} \left(\hat{a}^\dagger(\mathbf{p}_{k_s}, \mathbf{k}_s) \right)^2 / \sqrt{2} |\text{vac}\rangle, \quad (14)$$

where \mathbf{p}_{k_s} is a unit vector corresponding to the direction of oscillations of the vector \mathbf{D} of the SPDC radiation having a wave vector \mathbf{k}_s ; $\hat{a}^\dagger(\mathbf{p}_{k_s}, \mathbf{k}_s)$ is the bosonic operator of photon generation in a mode with \mathbf{p}_{k_s} polarization and wave vector \mathbf{k}_s ; $|\text{vac}\rangle$ is the vacuum state of the electromagnetic field. To achieve a quantum polarization state the SPDC only needs to determine the direction of polarization of its own wave propagating in a birefringent crystal.

In collinear geometry, the SPDC is $\mathbf{k}_s = \mathbf{k}_i \parallel \mathbf{k}_p$. The direction of the pump wave vector \mathbf{k}_p is given by two crystallographic angles, the polar angle θ_p and the azimuthal angle ϕ_p (Fig. 1). The coordinates of the vector \mathbf{k}_s in the coordinate system $\{X, Y, Z\}$

$$\mathbf{k}_s = |\mathbf{k}_s| \begin{pmatrix} \sin\theta_p \cos\phi_p \\ \sin\theta_p \sin\phi_p \\ \cos\theta_p \end{pmatrix}. \quad (15)$$

in the coordinate system \mathbf{f}_1 and \mathbf{f}_2 take values

$$\begin{aligned} \mathbf{f}_1 &= \sqrt{\frac{\epsilon_x \epsilon_y}{\cos^2 \phi_p \epsilon_x + \sin^2 \phi_p \epsilon_y}} \begin{pmatrix} \sin \phi_p \\ -\cos \phi_p \\ 0 \end{pmatrix}, \\ \mathbf{f}_2 &= (\epsilon_z)^{1/2} (\epsilon_x \cos^2 \phi_p + \epsilon_y \sin^2 \phi_p)^{-1/2} \times \\ &\times (\epsilon_x \cos^2 \phi_p \sin^2 \theta_p + \epsilon_y \sin^2 \phi_p \sin^2 \theta_p + \epsilon_z \cos^2 \theta_p)^{-1/2} \times \\ &\times \begin{pmatrix} \cos \theta_p \cos \phi_p \epsilon_x \\ \cos \theta_p \sin \phi_p \epsilon_y \\ -\sin \theta_p (\cos^2 \phi_p \epsilon_x + \sin^2 \phi_p \epsilon_y) \end{pmatrix}. \end{aligned} \quad (16)$$

Let's also switch to the coordinate system $\{x, y, z\}$, in which the z axis is directed along the vector \mathbf{k} , the x axis lies in the plane (Z, \mathbf{k}) . The direction of the vector oscillations \mathbf{D} will lie in the xy plane. The coordinate system $\{xyz\}$ will be called the coordinate system associated with pumping. The transformation of the coordinate system $\{X, Y, Z\} \rightarrow \{x, y, z\}$ can be performed using a sequence of rotations around the Z axis by an angle ϕ_p and then around the new y' axis by the angle θ_p . As a result, the transformation matrix of the coordinate system has the form

$$\begin{aligned} M &= R_y(-\theta_p) R_z(-\phi_p) = \\ &= \begin{pmatrix} \cos \theta_p \cos \phi_p & \cos \theta_p \sin \phi_p & -\sin \theta_p \\ -\sin \phi_p & \cos \phi_p & 0 \\ \cos \phi_p \sin \theta_p & \sin \theta_p \sin \phi_p & \cos \theta_p \end{pmatrix}, \end{aligned} \quad (17)$$

where

$$R_y(\theta) = \begin{pmatrix} \cos \theta & 0 & \sin \theta \\ 0 & 1 & 0 \\ -\sin \theta & 0 & \cos \theta \end{pmatrix}$$

is the rotation matrix around the y axis by an angle θ , and

$$R_z(\phi) = \begin{pmatrix} \cos \phi & -\sin \phi & 0 \\ \sin \phi & \cos \phi & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad (18)$$

is the rotation matrix around the z axis by an angle ϕ [40].

The vectors \mathbf{f}_1 and \mathbf{f}_2 in this coordinate system are written as

$$\mathbf{f}_1^{xyz} = \sqrt{\frac{\epsilon_x \epsilon_y}{\cos^2 \phi_p \epsilon_x + \sin^2 \phi_p \epsilon_y}} \begin{pmatrix} 0 \\ -1 \\ 0 \end{pmatrix},$$

$$\mathbf{f}_2^{xyz} = (\epsilon_z)^{1/2} (\epsilon_x \cos^2 \phi_p + \epsilon_y \sin^2 \phi_p)^{-1/2} \times$$

$$\times (\epsilon_x \cos^2 \phi_p \sin^2 \theta_p + \epsilon_y \sin^2 \phi_p \sin^2 \theta_p + \epsilon_z \cos^2 \theta_p)^{-1/2} \times$$

$$\times \begin{pmatrix} \frac{\epsilon_x + \epsilon_y}{2} + \frac{\epsilon_x - \epsilon_y}{2} \cos 2\phi_p \\ -\frac{\epsilon_x - \epsilon_y}{2} \cos \theta_p \cos 2\phi_p \\ 0 \end{pmatrix}. \quad (19)$$

If $\epsilon_x \neq \epsilon_y$, then to find the direction of oscillation of the vector \mathbf{D} of the related waves in an anisotropic medium, expressions for \mathbf{f}_1^{xyz} and \mathbf{f}_2^{xyz} from (19) should be inserted into (12) and thus get the values of the parameters s_1 and s_2 . At the same time, the directions of the vector oscillations \mathbf{D} , \mathbf{p}_1 and \mathbf{p}_2 are defined by the expressions

$$\begin{aligned} \mathbf{p}_1 &= \mathbf{f}_1^{xyz} \cos s_1 + \mathbf{f}_2^{xyz} \sin s_1, \\ \mathbf{p}_2 &= \mathbf{f}_1^{xyz} \cos s_2 + \mathbf{f}_2^{xyz} \sin s_2, \end{aligned} \quad (20)$$

and the refractive indices of the waves are equal respectively to $|\mathbf{p}_1|$ and $|\mathbf{p}_2|$.

For the case of uniaxial crystals, the expressions are simplified. Let's assume that

$$\epsilon_x = \epsilon_y = \epsilon_\perp, \quad \epsilon_z = \epsilon_\parallel.$$

Note that

$$(\mathbf{f}_1^{xyz}, \mathbf{f}_2^{xyz}) = 0,$$

and this means that

$$\mathbf{p}_1 = \mathbf{f}_1^{xyz}, \quad \mathbf{p}_2 = \mathbf{f}_2^{xyz}.$$

In this case

$$\mathbf{p}_1^u = \sqrt{\epsilon_\perp} \begin{pmatrix} 0 \\ -1 \\ 0 \end{pmatrix},$$

$$\mathbf{p}_2^u = \sqrt{\frac{\epsilon_\parallel \epsilon_\perp}{\epsilon_\perp \sin^2 \theta_p + \epsilon_\parallel \cos^2 \theta_p}} \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, \quad (21)$$

that is, we obtain known expressions for the refractive index of an *ordinary* wave

$$n_o = \sqrt{\epsilon_\perp}$$

and the *extraordinary* wave

$$n_e = (\sin^2 \theta_p / \epsilon_\perp + \cos^2 \theta_p / \epsilon_\parallel)^{-1/2}.$$

To find the direction of the vector \mathbf{D} , the concept of the main plane containing the optical axis of the crystal (Z) and the vector \mathbf{k} is usually introduced. As it is easy to see, the vector \mathbf{D} of an ordinary wave \mathbf{p}_1 is normal to the main plane, and the vector \mathbf{D} of an extraordinary wave \mathbf{p}_2 lies in the main plane.

The value of \mathbf{p}_1 or \mathbf{p}_2 , which corresponds to a large value of the refractive index, after normalization by a unit length, is the desired vector \mathbf{p}_{ks} in (14).

3.2. Non-collinear scattering mode

In the non-collinear SPDC mode, photons are generated in different modes. Such a regime is not degenerated, and the quantum polarization state of the SPDC radiation in the approximation of a given field of a plane monochromatic pump and an infinitely long crystal [41] can be represented as

$$|\psi\rangle \propto \int_0^\pi d\phi f(\phi) |\mathbf{p}_s(\theta_s(\phi), \phi)\rangle \otimes |\mathbf{p}_i(\theta_i(\phi), \phi + \pi)\rangle. \quad (22)$$

Here $f(\phi)$ is a function describing the dependence of the scattering efficiency on the azimuth angle, $|\mathbf{p}_{s,i}\rangle$ is the quantum polarization state of the photon in the signal (s) and idle (i) modes, having the form

$$|\mathbf{p}_{s,i}\rangle = p_{k_{s,i}}(\theta_{s,i}, \phi_{s,i}) \hat{a}^\dagger(p_{k_{s,i}}, k_{s,i}) |\text{vac}\rangle, \quad (23)$$

where $\mathbf{p}_{k_{s,i}}(\theta_{s,i}, \phi_{s,i})$ is a unit vector specifying the direction of oscillation of the SPDC radiation vector \mathbf{D} , scattered at the polar angle $\theta_{s,i}$ and the azimuthal angle $\phi_{s,i}$ (see Fig. 1), $\hat{a}^\dagger(\mathbf{p}_{k_{s,i}}, \mathbf{k}_{s,i})$ is a bosonic photon generation operator in a mode with polarization $\mathbf{p}_{k_{s,i}}$ and wave vector $\mathbf{k}_{s,i}$. The indices « s » and « i » correspond to the signal and idle waves respectively.

To find the direction of oscillation of the \mathbf{D} emission vector in the signal \mathbf{p}_{k_s} and idle \mathbf{p}_{k_i} waves in the coordinate system $\{XYZ\}$, it is enough to find the coordinates of the wave vectors \mathbf{k}_s and \mathbf{k}_i . This can be done using a sequence of four rotations (Fig. 1), combining unit vectors along Z in the coordinate system XYZ with a unit vector, directed along $\mathbf{k}_{s,i}$:

$$\mathbf{k}_{s,i} = |\mathbf{k}_{s,i}| R_z(\phi_p) R_y(\theta_p) R_z(\phi_{s,i}) R_y(\theta_{s,i}) \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}, \quad (24)$$

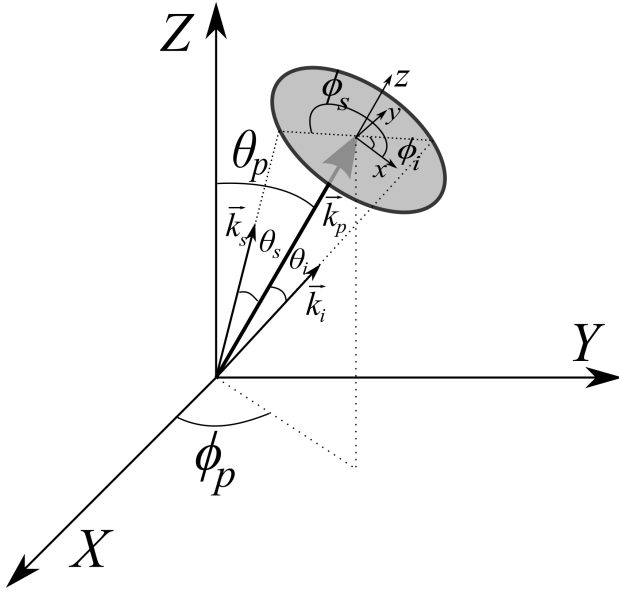


Fig. 1. Geometry of the SPDC with respect to the crystal-physical coordinate system $\{XYZ\}$ and the coordinate system $\{xyz\}$ associated with pumping.

where R_y and R_z are defined in (17), (18). Substituting (24) and (12) into (11) and taking the semi-axis with a greater length, we obtain the direction of the vector \mathbf{D} of the SPDC radiation necessary for calculating the quantum state (22).

3.3. Numerical calculations

According to the above expressions, the direction of the vector \mathbf{D} in the crystals BBO (barium β -borate) and BiBO (bismuth triborate) was calculated. For calculations, the Sellmeier equations for BBO [42] and BiBO were used (with correction for the refractive index of air, see [43]). Calculations were performed for pumping at a wavelength of 405 nm, in a frequency-adjusted mode. Scattering angles were considered 0, 3°, 10°, 17° and 30° outside the crystal, for a biaxial BiBO crystal, the scattering angle was considered for $\phi_s = 0$. For the BiBO crystal, calculations were performed for $\phi_p = 45^\circ$ and $\phi_p = 90^\circ$. Calculations for a uniaxial BBO crystal do not depend on the angle ϕ_p due to the symmetry of the ellipsoid of wave norms. The corresponding angles θ_p are shown in the table. The calculation of the polarization deviation angle of the SPDC is carried out, $\gamma(\theta_{s,i}, \phi_{s,i})$, defined as the angle between the directions the oscillation of the radiation vector \mathbf{D} in the non-collinear mode and the direction of the oscillation of the vector of the \mathbf{D} wave of the same type with $\mathbf{k}_{s,i} \parallel \mathbf{k}_p$ at the same parameters θ_p , ϕ_p and wavelength.

Table 1. Crystal parameters used in the calculation. Calculations were performed for BBO and BiBO crystals (with a fixed value of $\phi_p = 45^\circ$ and $\phi_p = 90^\circ$) with different values of θ_p , corresponding to the values of the scattering angle θ_s^{out} (0, 3°, 10°, 17° and 30°) outside the crystal when $\phi_s = 0$.

| | θ_s^{out} , deg. | θ_p , deg. | $\arccos s_1$, deg. | $\arccos s_2$, deg. |
|------------------------------|----------------------------|----------------------|-------------------------|-------------------------|
| BBO | 0 | 28.82 | 90.0 | 90.0 |
| | 3 | 29.24 | 86.8 | 86.8 |
| | 10 | 33.32 | 81.0 | 81.0 |
| | 17 | 41.15 | 78.6 | 78.6 |
| | 30 | 67.86 | 83.0 | 83.0 |
| BiBO, $\phi_p = 45^\circ$ | 0 | 141.39 | 90.0 | 90.0 |
| | 3 | 141.06 | 89.4 | 89.2 |
| | 10 | 137.74 | 88.4 | 86.8 |
| | 17 | 131.09 | 87.4 | 84.6 |
| | 30 | 104.84 | 88.8 | 87.5 |
| BiBO, $\phi_p = 90^\circ$ | 0 | 152.08 | 90 | 90 |
| | 3 | 151.71 | 88.5 | 88.5 |
| | 10 | 148.06 | 85.2 | 85.2 |
| | 17 | 141.05 | 82.8 | 82.8 |
| | 30 | 118.38 | 83.5 | 83.5 |

Note that in a uniaxial BBO crystal there are directions ($\phi_s = 0.180^\circ$) at which the polarization deviation angle is zero. This is due to the fact that the BBO crystal is uniaxial and negative, as a result of which the waves generated as a result of SPDC are ordinary. Under these conditions, the direction of the vectors \mathbf{D} is perpendicular to the same main plane. In a biaxial BiBO crystal, on the contrary, in a non-collinear mode, the direction of the vector \mathbf{D} does not coincide with the direction-using the vector \mathbf{D} in collinear mode.

Fig. 2 shows the dependences on the azimuthal scattering direction ϕ_s of the angle γ of the polarization deviation of the SPDC during the transition from the collinear to the non-collinear mode and the angle δ between the vectors \mathbf{D} of the signal and idle SPDC waves. In Fig. 2, it is seen that the value of the polarization deviation angle can take values exceeding 15°, while the value of the angle between the vectors \mathbf{D} of the signal and idle waves exceeds 30°.

It is also noteworthy that the graphs for BBO and BiBO at $\phi_p = 90^\circ$ are symmetric at $\phi_s \leftrightarrow 360^\circ - \phi_s$, which is due to the symmetry of the ellipsoid sections of the wave normal lines. At $\phi_p = 45^\circ$, this symmetry is lost, which also affects the symmetry of the direction of the vector \mathbf{D} .

In a uniaxial BBO crystal, the SPDC radiation is produced by an ordinary wave, and the value of the refractive index does not depend on the direction of scattering. As a result, the conditions of phase synchronization mean that the SPDC radiation forms a cone with a constant angle

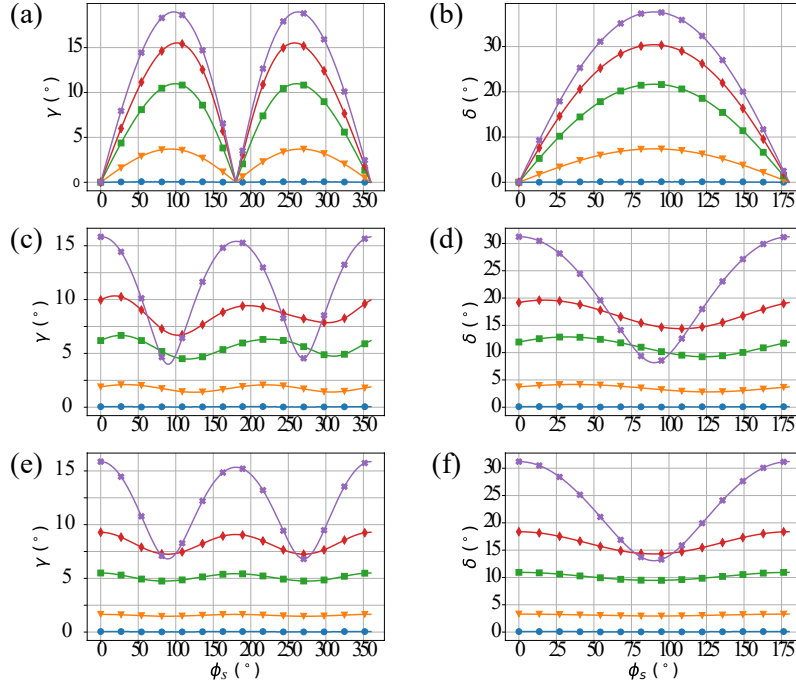


Fig. 2. *a, c, e* — Dependences of the angle γ between the vector \mathbf{D} of the SPDC radiation in non-collinear and collinear modes from the azimuthal scattering direction ϕ_s ; *b, d, f* are dependences of the angle δ between the vectors \mathbf{D} of the signal and idle waves on the azimuthal scattering direction ϕ_s . Graphs (*a, b*) are for the BBO crystal; (*c, d*) for the BiBO crystal, $\phi_p = 45^\circ$; (*e, f*) for BiBO crystal, $\phi_p = 90^\circ$. Blue circles, orange triangles, green squares, red diamonds and purple crosses correspond to the scattering angles $0^\circ, 3^\circ, 10^\circ, 17^\circ$ and 30° outside the crystal.

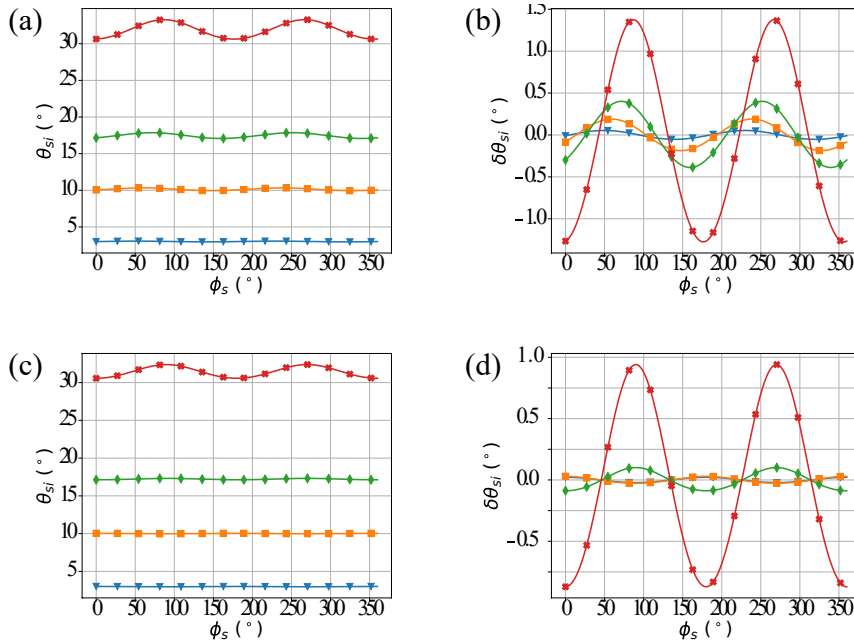


Fig. 3. *a, c* Dependences of the angle of non-collinearity (outside the crystal) on the azimuthal scattering direction ϕ_s ; *b, d* are dependences of the deviation of the scattering angle of the mean value in the azimuthal direction on the azimuthal scattering direction ϕ_s . Graphs (*a, b*) are for the BiBO crystal, $\phi_p = 45^\circ$; (*c, d*) for the BiBO crystal, $\phi_p = 90^\circ$. Blue triangles, orange squares, green diamonds, red crosses correspond to the scattering angles $3^\circ, 10^\circ, 17^\circ$ and 30° outside the crystal (at $\phi = 0$).

of solution (the angle of non-collinearity). This fact is widely known in the literature. As for the biaxial BiBO crystal, the refractive index for SPDC radiation depends on the scattering direction, and, in principle, the scattering angle may depend on the azimuthal scattering direction. No corresponding estimates have been made in the literature.

Fig. 3 shows the dependence of the angle of non-collimation (outside the crystal) on the azimuthal scattering direction for the BiBO crystal at $\varphi_p = 45^\circ$ and $\varphi_p = 90^\circ$. It can be seen from Fig. 3 that for scattering angles up to 17° , the value of the deviation of the scattering angle is relatively small. At the same time, at $\theta_s \approx 30^\circ$, the scattering angle varies within the range of about 2° , whereas for the case of $\varphi_p = 45^\circ$, the deviation is slightly greater than for $\varphi_p = 90^\circ$. Just like the direction of the vector \mathbf{D} , at $\varphi_p = 45^\circ$, unlike $\varphi_p = 90^\circ$, there is no symmetry $\varphi_s \leftrightarrow 360^\circ - \varphi_s$.

4. THE EFFECT OF SPDC POLARIZATION DEVIATION ON THE ENTANGLEMENT OF BIPHOTONS, GENERATED IN A TWO-CRYSTAL SCHEME

The phenomenon of polarization deviation in the non-collinear SPDC mode leads to a deterioration in the degree of quantum polarization entanglement of photonic pairs. One of the most well-known schemes for the generation of polarization-entangled photon pairs is the so-called two-crystal scheme (Fig. 4), consisting of two sequentially arranged identical nonlinear crystals oriented orthogonally. Falling on non-linear crystals, the pump is divided by polarization: its vertical component in the first crystal participates in the generation of photonic pairs, and the horizontal one does not participate and passes practically without interaction, since the conditions of phase synchronism for this component are not fulfilled. In the second crystal, on the contrary, there is a horizontal component that is involved in the generation of a photonic

The arrangement of crossed nonlinear crystals is shown pair, while the vertical pumping component f is not. In this case, the quantum state at the output of the two-crystal circuit in the approximation of plane signal, idle and pump waves, has the form [44]

$$|\Psi\rangle \propto \cos \varphi |\lambda_1\rangle \otimes |\lambda_2\rangle + \sin \varphi e^{i\phi} |\chi_1\rangle \otimes |\chi_2\rangle, \quad (25)$$

where the first term is the amplitude of the quantum polarization state generated in the first in a nonlinear crystal, and the second in the next nonlinear crystal, φ is the phase between the amplitudes, regulated by the ellipticity of the pump

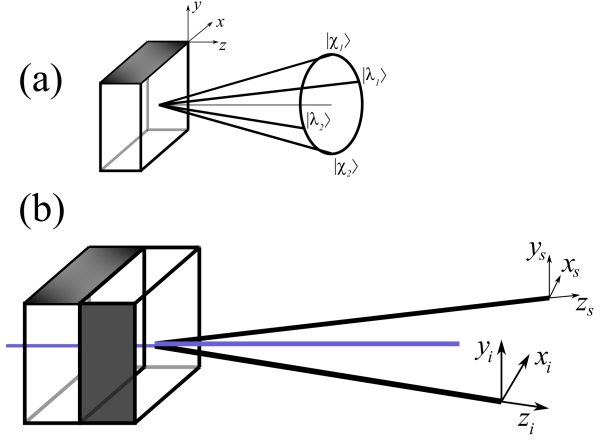


Fig. 4. (a) One of the crystals of the two-crystal scheme. The quantum polarization states of single photons are schematically shown corresponding to the collection directions in a two-crystal scheme. (b) The optical scheme of the source is a two-crystal scheme.

polarization, \otimes means the tensor product, φ the parameter describing the proportion of the quantum state generated in each of the nonlinear crystals in the quantum state $|\Psi\rangle$. State $|\lambda_1\rangle$ is the polarization state of a photon in the signal mode scattered at an azimuthal angle $\varphi_s = 0$, $|\lambda_2\rangle$ is the polarization state of a photon in the idle mode at $\varphi_i = 180^\circ$, $|\chi_1\rangle$ is the polarization state of a photon in the signal mode at $\varphi_s = 90^\circ$, $|\chi_2\rangle$ the polarization state of the photon at $\varphi_i = 270^\circ$. The dependence of the SPDC angle $\theta_{s,i}$ on the azimuthal direction is neglected. Each amplitude is the product of the polarization states of the photon in the signal and the idle modes, respectively. In the laboratory basis for the signal and idle states beam, the polarizations of a single photon have the form

$$\begin{aligned} |\lambda_1\rangle &= \begin{pmatrix} \cos \theta_1^s \\ \sin \theta_1^s \end{pmatrix}, |\lambda_2\rangle = \begin{pmatrix} \cos \theta_1^i \\ \sin \theta_1^i \end{pmatrix}, \\ |\chi_1\rangle &= \begin{pmatrix} \cos \theta_2^s \\ \sin \theta_2^s \end{pmatrix}, |\chi_2\rangle = \begin{pmatrix} \cos \theta_2^i \\ \sin \theta_2^i \end{pmatrix}, \end{aligned} \quad (26)$$

$\theta_{1,2}^{s,i}$ is the value of the angle between the direction of the vector \mathbf{D} and the unit vector $x_{s,i}$ in the signal and idle SPDC beams. The indices “1” and “2” correspond to the SPR radiation formed in the first and second crystals of the circuit, respectively. It is noteworthy that the polarization state of each photon is described in a two-dimensional complex Hilbert space, and the polarization state of a photon pair is described in a 2×2 dimension space. To describe the entanglement

in a two-particle system consisting of two-dimensional subsystems, there is a whole set of metrics [45], such as “Concurrence”, “Tangle” (coupling), entanglement of formation. We use Tangle because it is most sensitive to changes in the quantum state in the region of large degrees of quantum entanglement [46]. For a pure quantum state, entanglement can be expressed as [44]

$$T(|\Psi\rangle) = \left| \langle \Psi | \hat{\sigma}_2 \otimes \hat{\sigma}_2 | \Psi^* \rangle \right|, \quad (27)$$

where $*$ means complex conjugation, and

$$\hat{\sigma}_2 = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \quad (28)$$

is the Pauli matrix. Coupling takes the value of 1 for the maximally entangled quantum state and 0 for the factorized quantum state.

The entanglement for the quantum state (25) takes the value

$$T(|\Psi\rangle) = \frac{\sin^2 2\alpha (1 - |s_1|^2)(1 - |s_2|^2)}{1 + \sin 2\alpha \operatorname{Re}(s_1 s_2 e^{i\phi})}, \quad (29)$$

where

$$s_{1,2} = \langle \lambda_{1,2} | \chi_{1,2} \rangle.$$

The value of $T(|\Psi\rangle) = 1$, if $s_1 = s_2 = 0$, or

$$\begin{aligned} |s_1| &= |s_2|, \\ \alpha &= \pi/4 + n\pi, \\ \phi &= \pi - \arg(s_1 s_2) + n\pi, \end{aligned} \quad (30)$$

where n is an integer. The first condition means that the angles between the vectors $\mathbf{D} |\lambda_1\rangle, |\chi_1\rangle$ and $|\lambda_2\rangle, |\chi_1\rangle$ should be equal. The following condition means that the amplitude modules of the quantum state corresponding to the first and second crystals must be equal. The third condition says that to obtain a quantum state with the maximum possible degree of entanglement, it is necessary to set the optimal phase value between the radiation formed in the first and second crystals of the circuit.

Figure 5 shows the phase dependence of coupling ϕ for the BBO crystal and the crystal BiBO at $\phi_p = 45^\circ$ and $\phi_p = 90^\circ$. Calculations are performed for scattering angles $0^\circ, 3^\circ, 10^\circ, 17^\circ$ and 30° outside the crystal. It can be seen from Fig. 5 that for BiBO crystal at $\phi = 0$, with an increase in the scattering angle from 0 to 30° , the coupling decreases

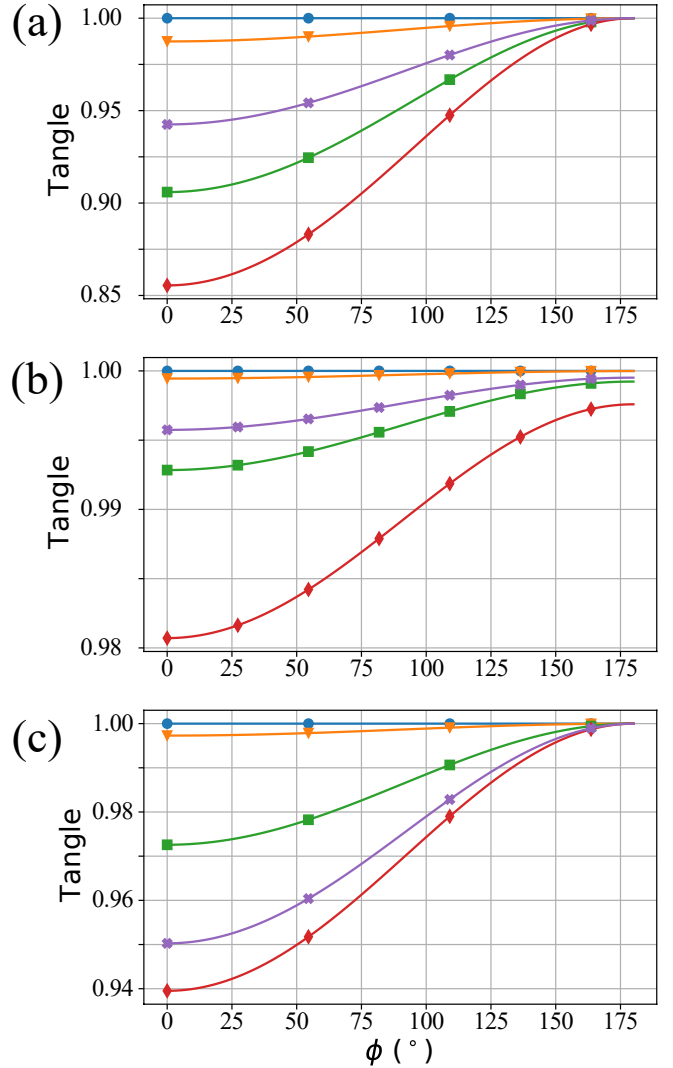


Fig. 5. Dependence of coupling (Tangle) on phase ϕ for BBO crystal (a), BiBO crystal at $\phi_p = 45^\circ$ (b), BiBO crystal at $\phi_p = 90^\circ$ (c). Blue circles, orange triangles, green squares, red diamonds and purple crosses correspond to the scattering angles $0^\circ, 3^\circ, 10^\circ, 17^\circ$ and 30° outside the crystal. The corresponding angles θ_p given in the table.

from 1 to 0.85. At the same time, for a biaxial BiBO crystal at $\phi_p = 45^\circ$, as the scattering angle increases, the coupling decreases by a smaller value (from 1 to 0.98). At $\phi_p = 90^\circ$, the decrease in cohesion is more pronounced compared to $\phi_p = 45^\circ$. At the same time, when using BBO crystals and BiBO crystals with $\phi_p = 90^\circ$, the coupling value can be completely restored to a value of 1 at $\phi = 180^\circ$. At $\phi_p = 45^\circ$, there is no complete recovery of coupling. This is due to the fact that due to the symmetry in the BBO crystal and the BiBO crystal $|s_1| = |s_2|$, and in a BiBO crystal at $\phi_p = 45^\circ$ $|s_1| \neq |s_2|$ (see the table).

5. CONCLUSION

In the paper, analytical expressions are given for the direction of oscillations of the vectors \mathbf{D} and \mathbf{E} of radiation propagating in uniaxial and biaxial nonlinear optical crystals, as well as the value of the refractive index. The obtained results were used to calculate the value of the polarization deviation angle of the SPDC γ , as well as the angle between the polarization of the signal and idle waves δ in BBO and BiBO crystals. It is shown that the value of γ can exceed 15° , and the value of γ is 30° . The obtained estimates indicate the importance of taking into account the deviation of the polarization of the SPDC in the non-collinear mode when creating sources of polarization-entangled photon pairs. Also, taking into account the polarization deviation of the SPDC is required when calculating the value of effective nonlinearity in a non-collinear SPDC. These results can also be used to create phantom polarimeters in which the systematic error due to the phenomenon of polarization deviation has been eliminated.

For the first time, estimates were made for the deviation of the cone shape of the SPDC. It is shown that at the scattering angle $\approx 30^\circ$ when the azimuthal direction changes, the scattering angle changes within 2° . At the same time, with the direction $\varphi_s = 0$; the 180° scattering angle decreases, and with $\varphi_s = 90^\circ$; the 270° scattering angle on the contrary increases. This means that in a two-crystal scheme with large scattering angles, an estimate of the deviation of the cone shape of the SPDC is required in order for the radiation generated in two sequentially arranged nonlinear crystals to spatially coincide.

It is found that the negative effect of polarization deviation on the degree of quantum entanglement (cohesion) in a two-crystal scheme when using biaxial crystals can be improved by choosing the optimal parameter φ , determined by the ellipticity of the pump polarization. At the same time, at $|s_1| = |s_2|$, the maximum degree of quantum entanglement can be achieved, and at $|s_1| \neq |s_2|$, the entanglement can only be partially restored. This result will make it possible in the future to create sources of polarization-entangled photon pairs with a degree of entanglement close to the maximum.

ACKNOWLEDGMENTS

The authors thank A.S. Chirkin for useful discussions and invaluable support-support at all stages of work.

FUNDING

The work was carried out at the expense of a grant from the Russian Science Foundation (project No.21-12-00155).

REFERENCES

1. D. N. Klyshko, JETP Lett. **6**, 490 (1967)
2. C. Zhang et al., Adv. Quant. Technol. **4**, 2000132 (2021).
3. S. V. Vintskevich, D. A. Grigoriev, and M. V. Fedorov, Laser Phys. Lett. **16**, 065203 (2019).
4. G. Brida, M. Genovese, and M. Gramegna, Laser Phys. Lett. **3**, 115 (2005).
5. A. N. Penin and A. V. Sergienko, Appl. Opt. **30**, 3582 (1991).
6. P. P. Gostev, D. P. Agapov, A. V. Demin et al., Measurement Techniques **61**, 1166 (2019)
7. P. A. Prudkovskii, P. A. Safronenkov, and G. Kh. Kitaeva, Opt. Lett. **47**, 4842 (2022).
8. J. Matthews, X.-Q. Zhou, H. Cable et al., NPJ Quant. Inf. **2**, 1 (2016).
9. C. Couteau, Contemp. Phys. **59**, 291 (2018).
10. D. Bouwmeester, J.-W. Pan, M. Daniell et al., Phys. Rev. Lett. **82**, 1345 (1999).
11. H.-S. Zhong, Y. Li, W. Li et al., Phys. Rev. Lett. **121**, 250505 (2018).
12. P.-G. Kwiat, E. Waks, and A. G. White, Phys. Rev. A **60**, R773 (1999).
13. C. E. Kuklewicz, M. Fiorentino, G. Messin et al., Phys. Rev. A **69**, 013807 (2004).
14. F. N. C. Wong, J. H. Shapiro, and T. Kim, Laser Phys. **16**, 1517 (2006).
15. M. Barbieri, C. Cinelli, F. de Martini et al., Laser Phys. **16**, 1439 (2006).
16. K. A. Kuznetsov, E. I. Malkova, and R. V. Zakharov, Phys. Rev. A **101**, 053843 (2020).
17. K. G. Katamadze and S. P. Kulik, JETP **112**, 20 (2011)
18. N. A. Borshchevskaya, F. Just, K. G. Katamadze et al., Laser Phys. Lett. **16**, 085207 (2019).
19. M. V. Chekhova and O. A. Shumilkina, JETP Lett. **91**, 649 (2010)
20. R. Rangarajan, L. E. Vicent, A. B. U'Ren, and P. G. Kwiat, J. Mod. Opt. **58**, 318 (2011).
21. M. V. Fedorov, Phys. Rev. A **93**, 033830 (2016).
22. M. Reichert, H. Defienne, and J. W. Fleischer, Scientific Reports **8**, 7925 (2018).
23. F. Just, A. Cavanna, M. V. Chekhova, and G. Leuchs, New J. Phys. **15**, 083015 (2013).
24. D. N. Frolovstev and S. A. Magnitskiy, Phys. Wave Phenomena **25**, 180 (2017).

25. D. N. Frolovtssev and S. A. Magnitskiy, EPJ Web of Conf. **220**, 03016 (2019).
26. A. Migdall, JOSA B **14**, 1093 (1997)
27. D. Yu. Stepanov, V. D. Shigorin, and G. P. Shipulo, Sov. J. Quant. Electron. **14**, 1315 (1984)
28. J. Q. Yao and T. S. Fahlen, J. Appl. Phys. **55**, 65 (1984).
29. N. Boeuf, D. A. Branning, I. Chaperot et al., Opt. Eng. **39**, 1016 (2000).
30. G.-W. Huo, T.-Y. Zhan, R.-G. Wan et al., Proc. SPIE **8333**, 261 (2012).
31. R. Akbari and Major, Laser Phys. **23**, 035401 (2013).
32. A. S. Chirkin, P. P. Gostev, D. P. Agapov, and S. A. Magnitskiy, Laser Phys. Lett. **15**, 115404 (2018).
33. S. A. Magnitskiy, D. P. Agapov, and A. S. Chirkin, Opt. Lett. **47**, 754 (2022).
34. D. A. Balakin and A. V. Belinsky, JETP **133**, 26 (2021)
35. L. D. Landau et al., Electrodynamics of Continuous Media Vol. 8, Elsevier (2013)
36. M. Born and E. Wolf, *Principles of Optics: Electromagnetic Theory of Propagation, Interference and Diffraction of Light*, Elsevier (2013)
37. V. G. Dmitriev and L. V. Tarasov, *Applied Nonlinear-Optics: Second-Harmonic Generators and Parametric Light-Generators*, Radio Sviaz, Moscow (1982)
38. E. Kreuzig, *Advanced Engineering Mathematics*, Willey (1972).
39. L. A. Krivitskii, S. P. Kulik, G. A. Maslennikov, and M. V. Chekhova, Quant. Electron. **35**, 69 (2005)
40. E. W. Weisstein, *Rotation Matrix*, Wolfram Research (2003).
41. L. Mandel and E. Wolf, *Optical Coherence and Quantum Optics*, Cambridge University Press, Cambridge (1995)
42. K. Kato, IEEE J. Quant. Electron. **22**, 1013 (1986).
43. H. Hellwig, J. Liebertz, and L. Bohat'y, J. Appl. Phys. **88**, 240 (2000).
44. D. N. Frolovtssev and S. A. Magnitskiy, Proc. of ICLO, **1** (2020).
45. W. K. Wothers, Quant. Inf. Comput. **1**, 27 (2001).
46. N. A. Peters, T.-C. Wei, and P. G. Kwiat, Phys. Rev. A **70**, 052309 (2004).