Monolithic symmetry effect on optimizing the phase matching parameters of a biaxial crystal for spontaneous parametric down conversion

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We report on the optimum phase matching parameters of collinear SPDC configuration for type I and type II in biaxial crystal. Considering the monolithic symmetry effect of BIBO, the double phase matching conditions based on angle-dependent refractive index are presented theoretically. Then we numerically simulate the effective nonlinear coefficient in all azimuth angles, and ascertain the maximal phase matching orientations. The results shows that calculations can be further simplified in one of the octant planes owing to monolithic symmetry of the crystal. It is superior in determination of phase matching conditions and the spatial distribution of entangled photons.

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1. Introduction

With rapid development of nonlinear optical material and technology, nonlinear crystals play an important role in laser generation, photoelectric devices, and image processing. It is generally classified as uniaxial and biaxial crystals according to birefringence phenomenon [1]. The optical properties, such as the velocities, propagation directions, polarization, and energy distribution, closely depend on crystal characters and incident light direction. By now nonlinear crystals have been widely applied in nonlinear frequency conversion, especially for the second harmonics generation (SHG). Since Kwiat *et al.* first demonstrated high efficiency polarization entanglement generation with BBO crystal in 1995 [2], the process of spontaneous parametric down-conversion (SPDC) in nonlinear crystals become the most accessible and controllable source of entanglement [3,4]. This SPDC based source of entangled photons that have strong correlations beyond what classical physics allows, has great potential applications in quantum information processing such as quantum teleportation [5,6], quantum computing and quantum swapping [7,8]. Nevertheless, the relatively low nonlinear optical coefficient of uniaxial crystal confines its application to a high-intensity source of multi-entangled photon pairs. Thus using biaxial crystals and periodically poled materials become a good option for brighter source of multi-entangled photons generation [9,10].

BiB_3O_6 (BIBO) is a newly developed monolithic negative biaxial crystal in past ten years. It has excellent optical characters such as exceptionally high nonlinearity coefficient, wide range of optical transparency and high damage threshold. Since BIBO was first introduced by Hellwig *et al.* [11,12], there are extensive researches on SHG in BIBO theoretically and experimentally [8,13-15]. Calculating the nonlinear frequency conversion in biaxial crystal is so difficult for its complex optical structure. Nearly all previous works of BIBO concentrated on double frequency process only [7,8,13-16], while the optimization parameters of phase matching conditions are considered in three principal planes with Yao's method [13,17]. It is attractive that BIBO crystal has been applied to generate multi-photon entanglement in quantum optics [18-20], owing to its high nonlinearity coefficient and biaxial nature of refractive-index distribution. However, the preceding approach is too complicated and cannot gain the angular derivative of refractive index in phase matching direction, which is a key parameter to influence the spatial distribution of entangled photons [21]. In virtue of the preeminent performance in nonlinear frequency conversion, there is an earnest need to investigate the angular derivative of refractive index and phase matching conditions in SPDC process with biaxial crystal. Working out a convenient method for determining the optimization phase matching parameters in SPDC process is meaningful.

In this paper, we show the optimum phase matching parameters of collinear SPDC configuration in biaxial crystal. Considering the monolithic symmetry effect of BIBO, we describe theoretically the double phase matching conditions for type I and type II, which is obtained by introducing the angle-dependent refractive index without solving the quadratic Fresnel equations. Then the effective nonlinear coefficient for two types phase matching is calculated in all azimuth angles, and the maximal phase matching orientations are ascertained. We emphasize on how to simplify the optimization phase matching condition based on the monolithic symmetry effect and angle-dependent refractive index. It is also superior in determination of the spatial distribution of entangled photons.

2. The double phase matching of BIBO crystal in SPDC process

We study double phase matching of BIBO crystal in collinear configuration SPDC as considering the angle dependent refractive indices. The value of angle dependent refractive indices for two linear polarized beams can be obtained from [1]

$$n_1 = \frac{n_x n_z}{\sqrt{n_z^2 \cos^2[(\theta_1+\theta_2)/2] + n_x^2 \sin^2[(\theta_1+\theta_2)/2]}}, \quad (1a)$$

$$n_2 = \frac{n_x n_z}{\sqrt{n_z^2 \cos^2[(\theta_1-\theta_2)/2] + n_x^2 \sin^2[(\theta_1-\theta_2)/2]}}. \quad 1b)$$

The angles θ_1, θ_2 are deduced by exploiting the angle definition introduced by Japanese mathematician Kodaira Kunihiko [17]. It can be expressed by [22]

$$\cos\theta_1 = (\cos\theta / \cos\gamma)\cdot\cos(\Omega-\gamma), \quad (2a)$$

$$\cos\theta_2 = (\cos\theta / \cos\gamma)\cdot\cos(\gamma+\Omega), \quad (2b)$$

here $\gamma = \arctan(\tan\theta\cdot\cos\phi)$ in the case of $0^\circ \le \theta < 90^\circ$, and θ is substituted by $\pi-\theta$ in the range of $90^\circ < \theta \le 180^\circ$. The optical axis angle $\Omega = \arcsin[\sqrt{(n_y^2-n_x^2)/(n_z^2-n_x^2)}\cdot n_z/n_y]$, is a function dependent on wavelength of pump waves. n_x, n_y, and n_z are the principal refractive indices at given wavelength. The schematic diagram in Fig.1 shows the coordinate transformation of the dielectric axis (xyz), the crystal principal axis (abc) and the lab frame ($x''y''z''$).For biaxial crystals in principal coordinates system (xyz), we designate the two optical axes as C_1, C_2 distributing symmetrically around z-axis, and the angles between z and C_1, C_2 are Ω, $-\Omega$, respectively. The orientation of wave vector $\vec{k}$ is expressed by (θ,ϕ), with θ representing the polar angle relative to z-axis and ϕ representing the azimuthal angle measured from x-axis in xy plane. It is obtained by first rotating the principal coordinates system (xyz) ϕ around z axis, acquired coordinates $x'y'z'$; and then the coordinate ($x'y'z'$) is rotated θ around y' axis, and gotten the lab frame coordinates $x''y''z''$. θ_1, θ_2 are angles between the wave vector $\vec{k}$ and C_1, C_2, and β is the angle between the crystal principal axis a and c.

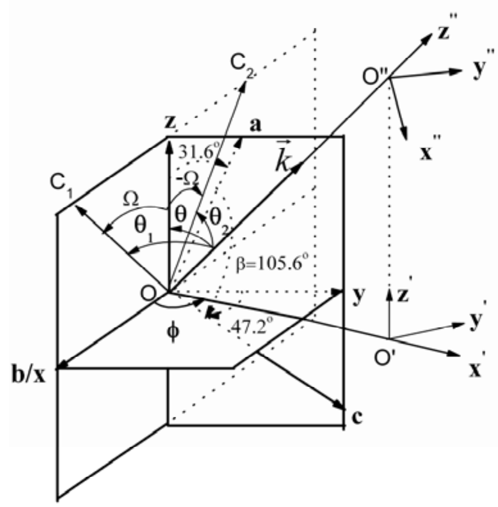

Fig. 1. The schematic diagram of biaxial crystal parameters in different coordinate system: the dielectric axis (xyz), the crystal principal axis (abc) and the lab frame ($x''y''z''$).

If the parameters Ω, θ and ϕ are known, we can acquire the value of angle dependent refractive indices. Generally, there are two different refractive indices in Eq.(1), and they are called the "fast" and the "slow", where the "fast" is with a smaller refractive index. The refractive indices in space distribution exert an influence on phase-matching conditions and birefringence anisotropy.

Phase matching is a vital element as other form of momentum and energy law in SPDC process. The incident photons are split into pairs of photons that have combined energies and momenta equal to the energy and momentum of the original photon. If the photons share the same polarization it is deemed the Type I, and if they have perpendicular polarizations it is called the Type II. Then the phase matching condition can be expressed by

$$\omega_p = \omega_s + \omega_i, \quad (3a)$$

$$\vec{k}_p = \vec{k}_s + \vec{k}_i, \quad (3b)$$

where ω_p, ω_s, ω_i are the frequency of the incident photon and two down-converted photons, and $\vec{k}_p$, $\vec{k}_s$, $\vec{k}_i$ are the corresponding wave vectors. The terms "signal" and "idler" are arbitrary. Since there are two refractive indices for each wavelength, the phase matching conditions are achieved in the following fashion:

$$\vec{k}_p(slow) = \vec{k}_s(fast) + \vec{k}_i(fast)\,, \qquad (4a)$$

$$\vec{k}_p(slow) = \vec{k}_s(fast) + \vec{k}_i(slow)\,. \qquad (4b)$$

Eq.(4a) are the type I phase matching configuration, and Eq.(4b) is the type II phase matching condition. Figure 2 shows the schematic diagram in bi-axial crystal, with the wave vector of pump light $\vec{k}_p$ in xz plane (the $\phi = 0$ plane). θ_j, ϕ_j (j=p,s,i) represent the corresponding polar angle and azimuthal angle of pump, signal and idler beam. The orientation of C is z axis. An auxiliary angle δ defines the polarization directions of the slow and fast waves (eigenmodes), which is between the polarization direction of the slow wave and the propagation plane containing z axis.

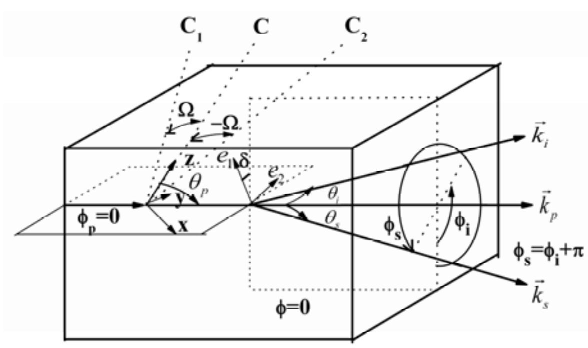

Fig. 2. The schematic diagram of SPDC process in biaxial crystal.

In the collinear degenerate case, $\omega_i = \omega_s = \omega_p/2$ and $\vec{k}_j = n_j(\theta_j,\phi_j)\omega_j / c \cdot \hat{k}_j\,(j = p,s,i)$, n_j is the refractive indices, and $\hat{k}_j$ is the unit vector with the same orientation. The polar angles of signal and idler beam are zero ($\theta_j = \theta_s = 0$), simultaneously. Then the type I and type II phase matching conditions can be expressed as follows:

$$2n_{ps}(\theta,\phi) = n_{sf}(\theta,\phi) + n_{if}(\theta,\phi)\,, \qquad (5a)$$

$$2n_{ps}(\theta,\phi) = n_{sf}(\theta,\phi) + n_{is}(\theta,\phi)\,, \qquad (5b)$$

Where n_{jm} (j = p, s, i; m=$slow$, $fast$) are the refractive indices of fundamental and SPDC waves. Substituting the refractive indices in Eq.(1) into Eq.(5), we can get the relation of phase matching angles (θ,ϕ) for the type I and type II in SPDC process. This is the foundation of considering the optimum phase matching orientation calculated by the effective nonlinear coefficient.

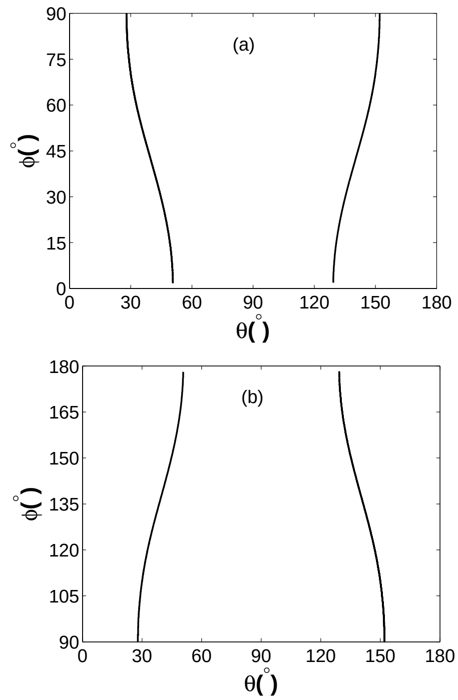

Fig. 3. The double phase matching conditions of type I phase-matching at a fundamental wavelength of 405 nm in SPDC process, (a) $\theta = 0^\circ \sim 180^\circ$, $\phi = 0^\circ \sim 90^\circ$ and *(b)* $\theta = 0^\circ \sim 180^\circ$, $\phi = 90^\circ \sim 180^\circ$.

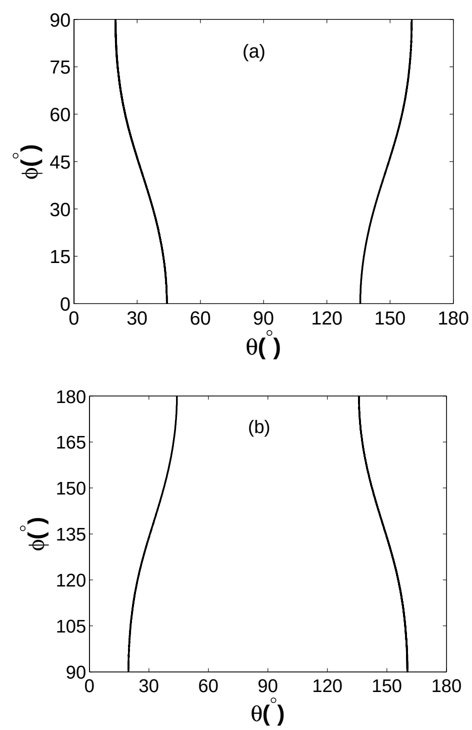

Fig. 4. The double phase matching conditions of type II phase matching at a fundamental wavelength of 780 nm in SPDC process, (a) $\theta = 0^\circ \sim 180^\circ$, $\phi = 0^\circ \sim 90^\circ$ *and (b)* $\theta = 0^\circ \sim 180^\circ$, $\phi = 90^\circ \sim 180^\circ$.

Owing to the monolithic symmetry of BIBO crystal, there are two possible phase matching directions at a given wavelength. We utilize the parameters from Hellwig's data at room temperature [23], and acquire the Sellmeier coefficients of dispersion relation at given wavelength. The relationships of phase matching angle (θ,ϕ) are numerically simulated with Eq.(1) and Eq.(5). Fig. 3 depicts the type I phase matching angles with a fundamental wavelength of 405nm, while Figure 4 is the type II phase-matching at wavelength 780nm. It is obvious that the curves distribute symmetrically with respect to θ=90° . The polar angle q is in the range of 27.9° £ q £ 50.7° and 129.3° £ q £ 152.1° for the type I, and it is 19.7° £ q £ 44.1° and 135.9° £ q £ 160.3° for the type II. It is also interesting to note that there can be two possible phase matching solutions for SPDC at angles θ and $180^\circ-\theta$, for a given azimuthal angle ϕ. These phenomena are generally called double phase matching conditions, and it is the monolithic symmetry result of the crystal. It is the fundament for analyzing on other parameters, such as the effective nonlinear coefficient d_{eff}.

3. The optimum phase matching orientations in BIBO crystal

The phase matching orientations have tremendous influence on conversion efficiency. We can calculate the general effective nonlinearity coefficients d_{eff} of the crystal to determine the optimum phase matching orientations. For the monolithic bi-axial crystal of point 2 group symmetry, the conversion efficiency depends on phase matching angle and the orientation of polarization. The expression of d_{eff} for the type I is given by [24]

$$\begin{aligned}
d_{eff}^{sff} = & -(d_{11}\cos^2\varphi+3d_{12}\sin^2\varphi)\cos^3\theta\cos\varphi\sin\delta\cos^3\delta \\
& -[d_{11}\sin^2\varphi+d_{12}(3\cos^2\varphi-2)]\cos\theta\cos\varphi\sin^3\delta \\
& +2[d_{11}\sin^2\varphi-d_{12}(3\sin^2\varphi-1)]\cos\theta\cos\varphi\sin\delta\cos^2\delta \\
& +2[d_{11}\cos^2\varphi-d_{12}(3\cos^2\varphi-1)]\cos^2\theta\sin\varphi\sin^2\delta\cos\delta \\
& -[d_{11}\cos^2\varphi-d_{12}(3\cos^2\varphi-1)]\cos^2\theta\sin\varphi\cos^3\delta \\
& -(d_{11}\sin^2\varphi+3d_{12}\cos^2\varphi)\sin\varphi\sin^2\delta\cos\delta \\
& -d_{13}\sin^2\theta\cos\delta[3\cos\theta\cos\varphi\sin\delta\cos\delta+\sin\varphi(3\cos^2\delta-2)] \\
& +d_{14}[\sin 2\theta\cos 2\varphi\cos\delta(3\sin^2\delta-1) \\
& +\sin\theta\sin 2\varphi\sin\delta(3\cos^2\theta\cos^2\delta+3\cos^2\delta-1)]
\end{aligned} , \qquad (6)$$

where d_{eff}^{sff} is the effective nonlinearity coefficient of type I, and d_{1j} (j=1,2,3,4) is the matrix tensor under the Kleinman symmetry condition [25].

For the type II phase-matching, the effective nonlinearity coefficient is expressed by [24]

$$\begin{aligned}
d_{eff}^{sfs} = & +(d_{11}\cos^2\varphi+3d_{12}\sin^2\varphi)\cos^3\theta\cos\varphi\sin^2\delta\cos\delta \\
& +[d_{11}\sin^2\varphi+d_{12}(3\cos^2\varphi-2)]\cos\theta\cos\varphi\cos^3\delta \\
& -2[d_{11}\sin^2\varphi-d_{12}(3\sin^2\varphi-1)]\cos\theta\cos\varphi\sin^2\delta\cos\delta \\
& +2[d_{11}\cos^2\varphi-d_{12}(3\cos^2\varphi-1)]\cos^2\theta\sin\varphi\sin\delta\cos^2\delta \\
& -[d_{11}\cos^2\varphi-d_{12}(3\cos^2\varphi-1)]\cos^2\theta\sin\varphi\sin^3\delta \\
& -(d_{11}\sin^2\varphi+3d_{12}\cos^2\varphi)\sin\varphi\sin\delta\cos^2\delta \\
& +d_{13}\sin^2\theta\sin\delta[3\cos\theta\cos\varphi\sin\delta\cos\delta-\sin\varphi(3\sin^2\delta-2)] \\
& +d_{14}[\sin 2\theta\cos 2\varphi\sin\delta(3\cos^2\delta-1) \\
& -\sin\theta\sin 2\varphi\cos\delta(3\cos^2\theta\sin^2\delta+3\sin^2\delta-1)]
\end{aligned} . \qquad (7)$$

We numerically simulate the effective nonlinear coefficient in Eq. (6) by considering the polarization orientation angle $\delta=0$. The normalized value of d_{j1} in BIBO are $d_{11}=2.53$ pm/V , $d_{12}=3.2$ pm/V , $d_{13}=-1.76$ pm/V , and $d_{14}=1.66$ pm/V . Combined with the double phase matching in Fig.3, the variation of the effective nonlinearity across the tuning range for type I phase matching configuration is shown in Fig. 5. The d_{eff} is calculated at a fundamental wavelength of 405nm, with the dash line presenting the case of $90^\circ<\theta<180^\circ$ and solid line corresponding to $0^\circ<\theta<90^\circ$. There are two different effective nonlinear coefficients for a given azimuthal angle ϕ at angles θ and $180^\circ-\theta$. The maximal absolute value of effective nonlinearity is 3.4869 pm/V at $\phi=-90^\circ$, q=27.9° (solid line) and $\phi=90^\circ$, q=152° (dashed line), making *yz* plane the most important one for nonlinear optical applications. It is obvious, that the effective nonlinear coefficient is distributed symmetrically around the axes $\phi=0^\circ$, $\phi=90^\circ$, and $\phi=-90^\circ$, due to the monolithic symmetry of BIBO, as shown in Figs. 5(a)-(d). So, it is advisable to consider the maximal phase matching orientations only in $0^\circ\le\phi\le 90^\circ$ range, just as in Fig. 5(a).

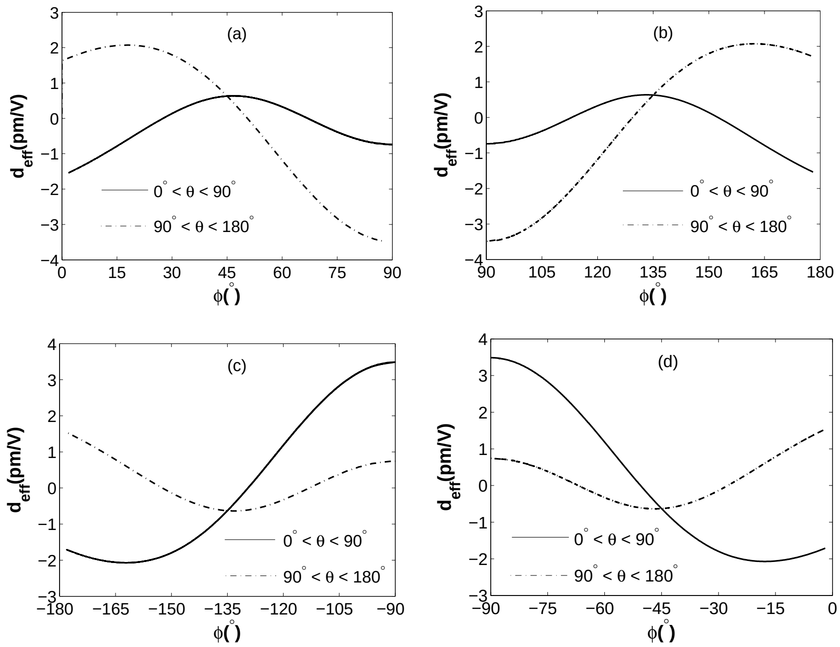

Fig. 5. The variation relation of d_{eff} along with ϕ for type I at fundamental wavelength of 405nm: (a) $\phi = 0^\circ \sim 90^\circ$, (b) $\phi = 90^\circ \sim 180^\circ$, (c) $\phi = -180^\circ \sim -90^\circ$, and (d) $\theta = -90^\circ \sim 0^\circ$.

Similar to the above process, we calculate Eq.(7) for δ =0 at a fundamental wavelength of 780nm. Considering the double phase matching condition in Fig.4, we obtain the d_{eff} varying with ϕ ($-180^\circ < \phi < 180^\circ$) for type II, as shown in Fig.6. The dash line represents the $90^\circ < \theta < 180^\circ$ case, while the solid line describes the case of $0^\circ < \theta < 90^\circ$. From the two directions in Fig.6, a same biggest absolute value of effective nonlinearity is 2.5323 pm/V at $\phi = 11.4^\circ$, q=43.1° and $\phi = 168.6^\circ$, q=43.1° in the case of $0^\circ < \theta < 90^\circ$, and at $\phi = -168.6^\circ$, q=136.9° and $\phi = -11.4^\circ$, q=136.9° for $90^\circ < \theta < 180^\circ$. Owing to afore mentioned monoclinic symmetry, the distributions of the effective nonlinear coefficient are symmetrical around the axes $\phi = 0^\circ$, $\phi = 90^\circ$, and $\phi = -90^\circ$, as shown in Figs. 6(a)-(d). Thus, taking into account the maximal phase matching orientations only in $0^\circ \le \phi \le 90^\circ$ range, as in Fig. 6(a), is a wise option.

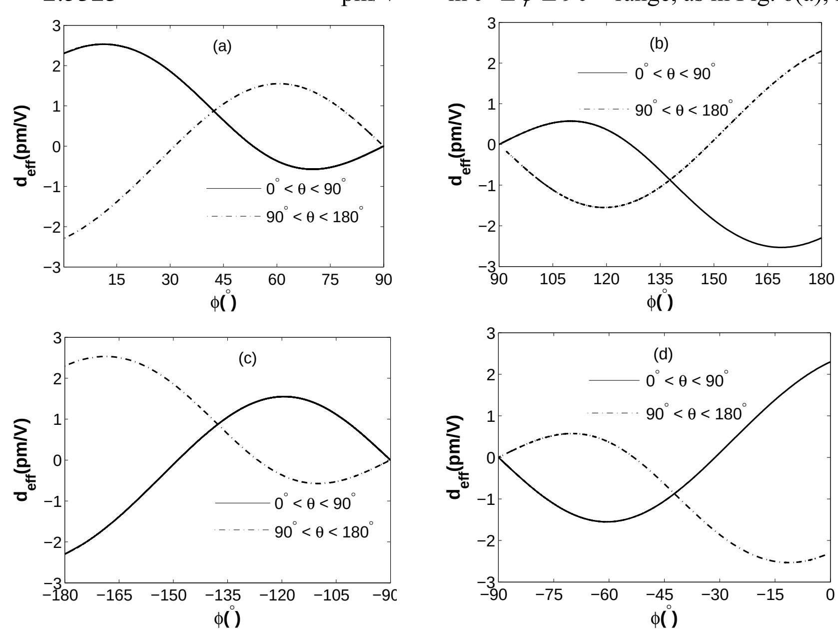

Fig. 6. The variation relation of d_{eff} along with ϕ for type II at fundamental wavelength of 780 nm: (a) $\phi = 0^\circ \sim 90^\circ$, (b) $\phi = 90^\circ \sim 180^\circ$, (c) $\phi = -180^\circ \sim -90^\circ$, and (d) $\theta = -90^\circ \sim 0^\circ$.

From the above analysis process, the monolithic symmetry is coming from the crystal axis ***a*** orthogonal to the plane containing crystal axes ***b*** and ***c***. Other symmetric effects are original in the analogy perpendicular relations of lines and planes. It makes the decision for the optimal phase matching conditions more convenient and simple to use in practice.

4. Conclusion

In conclusion, we have discussed in detail the phase matching conditions of collinear SPDC configuration in biaxial crystals. Based on angle-dependent refractive index and the monolithic symmetry effect of BIBO, we obtained double phase matching conditions for type I and type II, and then determined the optimum phase-matching directions by calculating the effective nonlinear coefficient. Our calculations show that there are several optimum phase matching parameters for two cases, while we can choose $\phi = -90^{\circ}$ for type I and $\phi = 11.4^{\circ}$ for type II only in case of $0^{\circ} < \theta < 90^{\circ}$ for simpleness due to the symmetry. This approach is convenient and accurate to avoid solving the quadratic Fresnel equations. It is superior in optimization of phase matching and determination of the spatial distribution of entangled photons.

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