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Nonlinear light beam propagation in uniaxial crystals: nonlinear refractive index, self-trapping and self-focusing

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Abstract. We derive the nonlinear paraxial wave equation for the propagation of an optical beam in nonlinear anisotropic media with centrosymmetry. As an application of the equation, we obtain the nonlinear refractive index (NRI) in three uniaxial crystals belonging to the symmetry classes 6/mmm of the hexagonal system, 4/mmm of the tetragonal system, and $\bar{3}$m of the trigonal system, respectively, and consider the self-trapping and self-focusing of the beam propagating in any direction in these crystals. We conclude that NRI, critical power and self-focusing length are all anisotropic (dependent upon the propagation direction) for an extraordinary light but isotropic for an ordinary light, and that there exists an elliptical self-trapping beam for the extraordinary light.

Keywords: Nonlinear refractive index, optical beam, self-focusing, self-trapping, uniaxial crystal

1. Introduction

A variety of nonlinear optical effects such as optical Kerr effects, ellipse rotations, self-focusings, self-phase modulations, and optical solitons, are related to the nonlinear refractive index (NRI) [1, 2], a phenomenon that refers to the intensity dependence on the refractive index, i.e. the refractive index $\bar{n}$ becomes

$$\bar{n} = n + n_2|E|^2,$$

where n is its linear part, E is an electric field, and n_2 is a NRI. The fundamental physical origin of the NRI is made clear via the formalism of nonlinear optical susceptibilities. The NRI n_2 is derived from the real part of a Fourier-transformed third-order susceptibility tensor $\chi^{(3)}$, and the specific linear combination of $\chi^{(3)}$ components which defines n_2 is dependent on the geometry. For linearly and circularly polarized light beams in isotropic materials, n_2 is related to only one component of $\chi^{(3)}$ [1, 2], but for beams linearly polarized at an angle relative to [100] and circularly polarized beams in the cubic crystals it becomes a little more complex, and is not isotropic as in the case of the linear refractive index [2, 3]. We can expect that, for crystals of lower symmetry, the linear combinations of $\chi^{(3)}$ components are generally more complex.

Being effects that result from the NRI, self-trapping and self-focusing of a light beam in isotropic Kerr media have been studied extensively for over three decades [4–9]. The circularly symmetric self-trapping beam in three dimensions was found as early as 1964 [4]. However, it is rather unstable. A small amount of deviation from the self-trapping solution will lead to either divergence or collapse [5, 6]. Moreover, if the power of the beam exceeds some critical power, the symmetric beam will self-focus to a point catastrophically [5–9]. Although a large amount of literature is available on the theory of self-trapping and self-focusing problems in isotropic media, few attempts have been made to deal with the case in anisotropic media because the latter is much more complicated than the former. To our knowledge, the first attempt to deal with the problem related to the optical Kerr effect in anisotropic media was made by Yumoto and Otsuka [10], but their paper was intended for a quasi-monochromatic plane wave propagating along a special direction (the major axes of the crystals) rather than the optical beam because the second-order spatial derivative of the field amplitude was not considered in their discussion. Karpman and Shagalov [11] considered self-focusing of the optical beam propagating parallel to an optic axis (c-axis) in uniaxial anisotropic gyrotropic media. In this paper we discuss the self-trapping and self-focusing of the light beam propagating along any direction in the uniaxial anisotropic media. In the following we will assume that the optical frequencies are small compared with the frequency of the fundamental electronic absorption of the material, but still large in relation to infrared vibrational frequencies. In this 'long-wavelength' limit, linear and nonlinear absorptions are negligible and Re $\chi^{(3)} \gg$ Im $\chi^{(3)}$.

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Structurally, the paper develops the thesis in the following way: in section 2 we shall obtain the expression of the light beam through the Taylor series expansion for the Fourier integral of an electric-field plane-wave spectrum vector about the transverse wavevector under the paraxial approximation condition, and derive the evolution equation of the beam, i.e. the nonlinear paraxial wave equation. The propagation coordinate system, defined as its z-coordinate axis coincident with the central wavevector of the beam, facilitates the derivation of the nonlinear paraxial wave equation. In section 3 we shall investigate the nature of $\chi^{(3)}$ under the coordinate transformation from the principal coordinate system to the propagation coordinate system, and obtain the nonlinear refractive index for three uniaxial crystals belonging to the symmetry classes 6/mmm of the hexagonal system, 4/mmm of the tetragonal system, and $\bar{3}$m of the trigonal system, respectively. In section 4 we shall consider the self-trapping and self-focusing of the beam propagating in any direction in these three crystals. Section 5, the final section, is set aside for conclusions. Appendix A deals with the proof of neglecting the terms that come from $\nabla\cdot\boldsymbol{E}$, and appendix B presents the calculations of the $\chi^{(3)}$ components under the transformation from the principal coordinate system to the propagation coordinate system.

2. Optical beam equation in anisotropic media

Assume that only one of the two mutually orthogonal eigenmodes is propagating in the transparent crystals, and under rather general conditions, a time-harmonic field propagating in the media is

$$\boldsymbol{E}(\boldsymbol{r},t) = \tfrac{1}{2}\boldsymbol{E}(\boldsymbol{r})\exp(-\mathrm{i}\omega t) + \text{c.c.},$$

where $\boldsymbol{E}(\boldsymbol{r})$ can be represented in a homogeneous half-space $z > 0$ by its plane-wave spectrum [12]

$$\boldsymbol{E}(\boldsymbol{r}) = \iint_{-\infty}^{\infty} \boldsymbol{E}_0(\boldsymbol{p})\exp[\mathrm{i}(\boldsymbol{K}\cdot\boldsymbol{R} + kz)]\,\mathrm{d}\boldsymbol{K}, \qquad (1)$$

where $\boldsymbol{E}_0$ represents the field of the normal wave, i.e. the plane wave, propagating along the wavevector $\boldsymbol{p}$, $\boldsymbol{p} = \boldsymbol{K} + k\boldsymbol{e}_z$, and $\boldsymbol{r} = \boldsymbol{R} + z\boldsymbol{e}_z$ ($\boldsymbol{e}_z$ is the unit vector along the z-coordinate, $\boldsymbol{K}$ and $\boldsymbol{R}$ represent transverse wavevector and coordinate vector perpendicular to $\boldsymbol{e}_z$, respectively, $\boldsymbol{K} = K_x\boldsymbol{e}_x + K_y\boldsymbol{e}_y$, $|\boldsymbol{e}_x| = |\boldsymbol{e}_y| = 1$). $\boldsymbol{E}_0$ and $\boldsymbol{p}$, sometimes refered to in the literature as the eigenfunction and eigenvalue, respectively, satisfy the eigenvalue equation [12,13]

$$\left(\boldsymbol{pp} - p^2\hat{I} + \frac{\omega^2}{c^2}\frac{\hat{\varepsilon}}{\varepsilon_0}\right)\cdot\boldsymbol{E}_0 = 0, \qquad (2)$$

where $\hat{I}$ and $\hat{\varepsilon}$ are unit and dielectric tensors of rank 2, respectively. k as a function of $\boldsymbol{K}$ can be derived from the dispersion equation

$$\det\left(\boldsymbol{pp} - p^2\hat{I} + \frac{\omega^2}{c^2}\frac{\hat{\varepsilon}}{\varepsilon_0}\right) = 0, \qquad (3)$$

where ω remains unchangeable.

Consider now the special case of a paraxial beam, which is a group of plane waves with the same frequency but slightly different directions of propagation. The wavevectors $\boldsymbol{p}$ of the component waves fill a solid angle around a central wavevector, that is the 'mean wavevector'. If we define the coordinate system, termed the propagation coordinate system hereinafter, such that its z-coordinate axis coincides with the central wavevector of the light beam, under the paraxial approximation which means $K/k \ll 1$, $\boldsymbol{E}_0(\boldsymbol{p})$ and k can be expanded in a Taylor series in the vicinity of the point $\boldsymbol{K} = 0$:

$$\boldsymbol{E}_0(\boldsymbol{p}) = \boldsymbol{E}_0^{(0)}(k_0) + \cdots, \qquad (4a)$$

$$k(\boldsymbol{K}) = k_0 + \nabla_K k_0\cdot\boldsymbol{K} + \tfrac{1}{2}\nabla_K\nabla_K k_0 : \boldsymbol{KK} + \cdots, \qquad (4b)$$

where $\boldsymbol{E}_0^{(0)} = \boldsymbol{E}_0(\boldsymbol{p})|_{K=0}$, $k_0 = k(\boldsymbol{K})|_{K=0}$ which is the magnitude of the central wavevector, $\nabla_K k_0 = \nabla_K k(\boldsymbol{K})|_{K=0}$, and so on, and $\nabla_K = \partial/\partial K_x\boldsymbol{e}_x + \partial/\partial K_y\boldsymbol{e}_y$. Substitution of the above expansion into equation (1) gives

$$\boldsymbol{E}(\boldsymbol{r}) = A(\boldsymbol{R},z)\boldsymbol{e}^0\exp(\mathrm{i}k_0z) + \mathrm{O}(K), \qquad (5)$$

where

$$A(\boldsymbol{R},z) = E_0^{(0)}\iint_{-\infty}^{\infty}\exp[\mathrm{i}\varphi(\boldsymbol{K},\boldsymbol{R},z)]\,\mathrm{d}\boldsymbol{K}, \qquad (6)$$

is the optical beam which is a scalar function, a phase factor φ reads

$$\varphi(\boldsymbol{K},\boldsymbol{R},z) = (\nabla_K k_0\cdot\boldsymbol{K} + \tfrac{1}{2}\nabla_K\nabla_K k_0 : \boldsymbol{KK})z + \boldsymbol{K}\cdot\boldsymbol{R},$$

and $\boldsymbol{e}^0$ is a unit vector of $\boldsymbol{E}_0^{(0)}$ that is the first-order approximation of the eigenfunction $\boldsymbol{E}_0$. Also, $\boldsymbol{E}_0^{(0)}$ should comply with equation (2) provided that expansion (4a) is only retained in the first term, that is,

$$(\boldsymbol{KK} - \boldsymbol{K}\cdot\boldsymbol{K}\hat{I})\cdot\boldsymbol{E}_0^{(0)} + k(\boldsymbol{K}\boldsymbol{e}_z + \boldsymbol{e}_z\boldsymbol{K})\cdot\boldsymbol{E}_0^{(0)} + k^2(\boldsymbol{e}_z\boldsymbol{e}_z - \hat{I})\cdot\boldsymbol{E}_0^{(0)} + \frac{\omega^2}{c^2\varepsilon_0}\hat{\varepsilon}\cdot\boldsymbol{E}_0^{(0)} = 0. \qquad (7)$$

In the following, we will discuss the evolution of the light beam (5) when a perturbed cubic nonlinearity appears in the media. Without loss of generality, we can assume that the media possess a centre of symmetry, some examples of which are [14] the ones belonging to the crystal classes 6m and 6/mmm in the hexagonal system, $\bar{3}$ and $\bar{3}$m in the trigonal system, and 4/m and 4/mmm in the tetragonal system. For these crystals, the lowest-order non-zero nonlinear susceptibility should be $\chi^{(3)}$ rather than $\chi^{(2)}$, therefore, the time-harmonic Maxwell equation in the mks system of units reads [13,15,16]

$$\nabla\times(\nabla\times\boldsymbol{E}) - \frac{\omega^2}{c^2\varepsilon_0}(\hat{\varepsilon}\cdot\boldsymbol{E} + \boldsymbol{P}_{\mathrm{NL}}) = 0, \qquad (8)$$

where $\boldsymbol{P}_{\mathrm{NL}}(\boldsymbol{r})$ expressed as

$$\boldsymbol{P}_{\mathrm{NL}}(\boldsymbol{r}) = \frac{3\varepsilon_0}{4}\chi^{(3)}(\omega = \omega + \omega - \omega) : \boldsymbol{E}(\boldsymbol{r})\boldsymbol{E}(\boldsymbol{r})\boldsymbol{E}^*(\boldsymbol{r}) \qquad (9)$$

is the third-order nonlinear polarization [14–16], and the fourth-rank tensor $\chi^{(3)}(\omega = \omega_1 + \omega_2 + \omega_3)$ is the Fourier transform of the third-order nonlinear susceptibility.

In order to obtain the wave equation for the slowly varying amplitude $A(\boldsymbol{R},z)$, the first step is to introduce equation (5) into (8) with the help of equation (7), and neglecting the second derivative $\partial^2 A/\partial z^2$ in the intermediate result. In this way, equation (8) can be deduced as

$$\iint_{-\infty}^{\infty} (k-k_0)(\boldsymbol{K}\boldsymbol{e}_z+\boldsymbol{e}_z\boldsymbol{K})\cdot\boldsymbol{E}_0^{(0)}\exp(\mathrm{i}\varphi)\,\mathrm{d}\boldsymbol{K} + (\nabla_\perp \boldsymbol{e}_z+\boldsymbol{e}_z\nabla_\perp)\cdot\boldsymbol{e}^0\frac{\partial A}{\partial z} + (\boldsymbol{e}_z\boldsymbol{e}_z-\hat{I}) \cdot\left[2\mathrm{i}k_0\boldsymbol{e}^0\frac{\partial A}{\partial z}+\boldsymbol{E}_0^{(0)}\iint_{-\infty}^{\infty}(k^2-k_0^2)\exp(\mathrm{i}\varphi)\,\mathrm{d}\boldsymbol{K}\right] - \frac{\omega^2}{c^2\varepsilon_0}\boldsymbol{P}_{\mathrm{NL}}\exp(-\mathrm{i}k_0 z)=0, \tag{10}$$

where a subscript $\perp$ represents the transverse part of a vector perpendicular to $\boldsymbol{e}_z$. The first two terms of the above equation result from $\nabla\cdot\boldsymbol{E}$, and as proved in appendix A, can be neglected. Retaining the first three terms of expansion (4*b*), we have

$$k^2-k_0^2\approx 2k_0(k-k_0)=2k_0\nabla_K k_0\cdot\boldsymbol{K}+k_0\nabla_K\nabla_K k_0:\boldsymbol{K}\boldsymbol{K}. \tag{11}$$

Finally, substituting equation (11) into (10) whose first two terms are neglected, we derive the optical beam equation in anisotropic media, i.e., the nonlinear paraxial wave equation:

$$\mathrm{i}\left(\frac{\partial A}{\partial z}-\nabla_K k_0\cdot\nabla_\perp A\right)-\frac{1}{2}\nabla_K\nabla_K k_0:\nabla_\perp\nabla_\perp A + \frac{\omega n_2}{c\alpha_1}|A|^2A=0, \tag{12}$$

where

$$\alpha_1=\boldsymbol{e}^0\cdot(\hat{I}-\boldsymbol{e}_z\boldsymbol{e}_z)\cdot\boldsymbol{e}^0=|e^0|_\perp, \tag{13}$$

and where the NRI n_2 is defined as

$$n_2=\frac{3\omega}{8ck_0}\chi^{(3)}:\boldsymbol{e}^0\boldsymbol{e}^0\boldsymbol{e}^0\boldsymbol{e}^0. \tag{14}$$

At this point it is important to emphasize the following. First, the field is determined by equation (5) in the lowest-order approximation. Since $\boldsymbol{E}(\boldsymbol{r})$ is proportional to $A(\boldsymbol{R},z)$, the vectorial field is completely specified when the scalar equation (12) is solved for $A(\boldsymbol{R},z)$. The reason why it is possible to describe the field as consisting of vectorial properties with the scalar equation can be readily elucidated through the physical background of the problem. As already pointed out, in this paper we limit ourselves to the discussion of the evolution of the light field under the influence of the perturbable nonlinearity. It is because the nonlinearity is perturbed that it can alter nothing more than the magnitude of the field rather than the direction of the field, which is governed by the dominant linear part of the field equation, i.e. equation (2) or equivalently equation (7). Moreover, because the nonlinearity has the property of a tensor, the vectorial property (polarization) of the field can, in turn, have an effect on the magnitude through the NIC n_2 within the last term of equation (12), and the NICs are different for the different kinds of polarization. Secondly, equation (12) is a general form of the evolution equation for the beam in nonlinear crystals. To find its concrete expression for the different modes, we should first obtain $\boldsymbol{E}_0^{(0)}$ and its accompanying function $k(\boldsymbol{K})$ by making use of the eigenvalue equation (7) and the dispersion equation (3), and then obtain k_0, $\nabla_K k_0$, $\nabla_K\nabla_K k_0$, α_1 and n_2. For example, in isotropic media with the linear refractive index $n=n_0$, the eigenfunction $\boldsymbol{E}_0^{(0)}$ is a linearly polarized vector field orthogonal to the wave propagation direction, and the dispersion equation becomes $K^2+k^2=\omega^2n_0^2/c^2$ such that $\alpha_1=1$, $k_0=\omega n_0/c$, $\nabla_K k_0=0$, and $\nabla_K\nabla_K k_0=-\hat{I}/k_0$. Suppose $\boldsymbol{E}_0^{(0)}$ is polarized along the x-axis; therefore, the NRI becomes

$$n_2=\frac{3}{8n_0}\chi^{(3)}_{xxxx}, \tag{15}$$

and the nonlinear paraxial wave equation is reduced to the previously derived (1+2)-dimensional nonlinear Schrödinger equation [5, 7–9]:

$$\mathrm{i}\frac{\partial A}{\partial z}+\frac{1}{2k_o}\left(\frac{\partial^2 A}{\partial x^2}+\frac{\partial^2 A}{\partial y^2}\right)+\frac{\omega n_2}{c}|A|^2A=0. \tag{16}$$

3. NRI in uniaxial crystals

For crystals, the problem becomes much more complicated than the case for the isotropic media because of the anisotropy of the linear refractive index and the fourth-rank tensor $\chi^{(3)}$. The nonlinear paraxial wave equation (12) is only derived under the condition of one-mode propagation. To ensure this condition is satisfied, we must investigate the nature of $\chi^{(3)}$ under coordinate transformation.

As mentioned above, equation (12) is obtained in the propagation coordinate system, but the allowed form of the third-order nonlinear susceptibility $\chi^{(3)}$ is determined in the principal coordinate system [13]. Therefore, $\chi^{(3)}$ should be transformed from the principal coordinate system (x',y',z'), where a prime represents the quantity in the principal coordinate system, to the propagation coordinate system (x,y,z). The corresponding new components $\chi^{(3)}_{ijkl}$ of the tensor $\chi^{(3)}$ can be computed in terms of the old ones $\chi^{(3)}_{i'j'k'l'}$ by writing [17]

$$\chi^{(3)}_{ijkl}=\sum_{i',j',k',l'}\chi^{(3)'}_{i'j'k'l'}g_{i'i}g_{j'j}g_{k'k}g_{l'l} \tag{17}$$

where $g_{i'i}=\boldsymbol{e}_i\cdot\boldsymbol{e}'_{i'}$ represents the direction cosine of the angle between the direction of the old base vector $\boldsymbol{e}'_{i'}$ and the new base vector $\boldsymbol{e}_i$. Because x' and y' directions are equivalent for the uniaxial crystals [13], we can thus choose the propagation coordinate system such that the z–x plane is within the z'–x' plane without loss of generality, as shown in figure 1. In this way, we can have

$$g_{x'x}=\cos(\theta),\quad g_{x'z}=\sin(\theta),\quad g_{z'x}=-\sin(\theta),\quad g_{z'z}=\cos(\theta),\quad g_{y'y}=1,\quad g_{x'y}=g_{y'x}=g_{y'z}=g_{z'y}=0, \tag{18}$$

where θ is the angle between the c-axis and the central wavevector of the beam, varying in a closed interval $[0,\pi]$. There are six classes of crystals with centrosymmetry: 6m and 6/mmm in the hexagonal system, $\bar{3}$ and $\bar{3}$m in the trigonal system, and 4/m and 4/mmm in the tetragonal system, respectively. Three of these crystals, 6/mmm, $\bar{3}$m and 4/mmm, whose $\chi^{(3)}$ form in the principal coordinate system can be found from table 1.5.2 of [14], can be easily demonstrated by the transformation above to have

$$\chi^{(3)}_{xyyy}=\chi^{(3)}_{yxxx}=\chi^{(3)}_{zyyy}=\chi^{(3)}_{yzzz}=0 \tag{19}$$

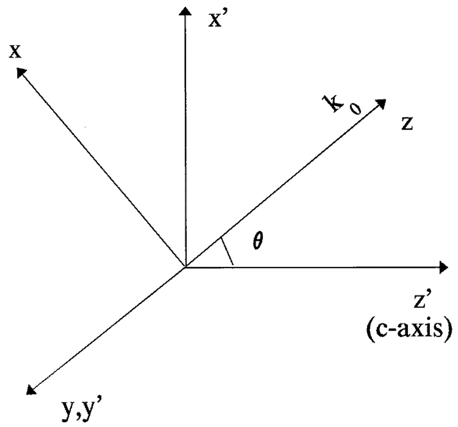

Figure 1. Coordinate systems and the central wavevector of the beam $\boldsymbol{k}_0$ that makes an angle θ with the optic axis ($0 \leqslant \theta \leqslant \pi$). The propagation coordinate system (x, y, z) is introduced by a counterclockwise rotation through θ about the y' axis of the principal coordinate system (x', y', z').

in the propagation coordinate system for any θ in the closed interval $[0, \pi]$. This means that if the field structure at $z = 0$ excites only one of the two orthogonal eigenmodes, there will not be a nonlinear couple between the o-light polarized along the y-axis and the e-light polarized in the z–x plane no matter which direction it propagates along: the mode merely keeps propagating in these three class crystals. But the other three centrosymmetric crystals belonging to crystal classes 6/m, $\bar{3}$ and 4/m can be proved not to have this feature.

It is this nonlinear couple-free feature of the o-light and the e-light at any direction that makes the discussion possible and simpler. Then, we can respectively deal with the evolution of the o-light and the e-light in the three class of crystals: $\bar{3}$m, 6/mmm and 4/mmm.

For the o-light propagating along any direction except the one parallel to the c-axis ($\theta = 0$ or π), the case is the same as that of isotropic media, as discussed at the end of section 2, and n_2 for the o-light is

$$n_2^{(\mathrm{o})} = \frac{3\chi^{(3)'}_{x'x'x'x'}}{8n_\mathrm{o}}. \tag{20}$$

When the central wavevector is along the direction parallel to the c-axis, there will be a special situation, which is a double mode with two eigenmodes, o-light and e-light, sharing the same dispersion properties, and the propagation becomes more complicated because of the singularity in this direction [13, 18]. This situation is not considered in this paper.

The propagation of the e-light is more complex than the case of the o-light.

The dispersion equation (3) for the e-light reads

$$\frac{K'^2_{x'} + K'^2_{y'}}{n_\mathrm{e}^2} + \frac{k'^2}{n_\mathrm{o}^2} - \frac{\omega^2}{c^2} = 0 \tag{21}$$

in the principal coordinate system, where n_e and n_o are the extraordinary index and ordinary index, respectively. Under the coordinate transformation shown in figure 1, the wavevector in the principal coordinate system, $\boldsymbol{p}'$, is related to the wavevector in the propagation coordinate system, $\boldsymbol{p}$, through

$$\begin{aligned} K'_{x'} &= K_x \cos\theta + k\sin\theta, \\ K'_{y'} &= K_y, \\ k' &= -K_x\sin\theta + k\cos\theta. \end{aligned} \tag{22}$$

Inserting equation (22) into (21) gives the dispersion equation in the propagation coordinate system:

$$\begin{aligned} &(n_\mathrm{o}^2\cos^2\theta + n_\mathrm{e}^2\sin^2\theta)K_x^2 + n_\mathrm{o}^2K_y^2 + (n_\mathrm{o}^2\sin^2\theta + n_\mathrm{e}^2\cos^2\theta)k^2 \\ &\quad + (n_\mathrm{o}^2 - n_\mathrm{e}^2)\sin(2\theta)K_xk - \frac{\omega^2n_\mathrm{e}^2n_\mathrm{o}^2}{c^2} = 0 \end{aligned} \tag{23}$$

for $0 < \theta < \pi$. The reason why θ is limited to the open interval $(0, \pi)$ is that, as mentioned previously, when the central wavevector is in the direction parallel to the c-axis, the o-light and the e-light will share the same dispersion properties. Therefore, after obtaining the dispersion equation, we can, respectively, have

$$\begin{aligned} &k_0 = \frac{\omega n^\mathrm{e}}{c}, \qquad (\nabla_Kk_0)_y = 0, \\ &(\nabla_Kk_0)_x = \frac{(n_\mathrm{e}^2 - n_\mathrm{o}^2)\sin(2\theta)}{2(n_\mathrm{e}^2\cos^2\theta + n_\mathrm{o}^2\sin^2\theta)}, \\ &(\nabla_K\nabla_Kk_0)_{xx} \\ &\quad = \frac{1}{k_0}\left\{-\frac{n_\mathrm{o}^2\cos^2\theta + n_\mathrm{e}^2\sin^2\theta}{n_\mathrm{e}^2\cos^2\theta + n_\mathrm{o}^2\sin^2\theta} + [(\nabla_Kk_0)_x]^2\right\}, \\ &(\nabla_K\nabla_Kk_0)_{yy} = -\frac{n_\mathrm{o}^2}{k_0(n_\mathrm{e}^2\cos^2\theta + n_\mathrm{o}^2\sin^2\theta)}, \\ &(\nabla_K\nabla_Kk_0)_{xy} = (\nabla_K\nabla_Kk_0)_{yx} = 0, \end{aligned} \tag{24}$$

where

$$n^\mathrm{e} = \frac{n_\mathrm{e}n_\mathrm{o}}{(n_\mathrm{e}^2\cos^2\theta + n_\mathrm{o}^2\sin^2\theta)^{1/2}} \tag{25}$$

is the linear refractive index for the e-light.

On the other hand, as is known [13], the electric displacement vector of the e-light is perpendicular to the central wavevector of the beam, but its electric field vector is, in general, not perpendicular to the central wavevector. The electric displacement vector and electric field vector form the angle δ, which is also the angle between the Poynting vector and the central wavevector, as shown in figure 2, and δ is given by [12, 18]

$$\delta = \arctan\frac{(n_\mathrm{e}^2 - n_\mathrm{o}^2)\sin(2\theta)}{2(n_\mathrm{e}^2\cos^2\theta + n_\mathrm{o}^2\sin^2\theta)}. \tag{26}$$

Then, the unit vector of the electric field $\boldsymbol{E}_0^{(0)}$ is found to be

$$\boldsymbol{e}^0 = \cos\delta\boldsymbol{e}_x + \sin\delta\boldsymbol{e}_z. \tag{27}$$

The combination of equation (14) with (27) gives the NRI in the propagation coordinate system

$$\begin{aligned} n_2(\theta) = \frac{3}{8n^\mathrm{e}}[&\cos^4\delta\cdot\chi^{(3)}_{xxxx} \\ &+ \cos^3\delta\sin\delta(\chi^{(3)}_{zxxx} + \chi^{(3)}_{xzxx} + \chi^{(3)}_{xxzx} + \chi^{(3)}_{xxxz}) \\ &+ \cos^2\delta\sin^2\delta(\chi^{(3)}_{zzxx} + \chi^{(3)}_{zxzx} \\ &+ \chi^{(3)}_{zxxz} + \chi^{(3)}_{xzzx} + \chi^{(3)}_{xzxz} + \chi^{(3)}_{xxzz}) \\ &+ \cos\delta\sin^3\delta(\chi^{(3)}_{xzzz} + \chi^{(3)}_{zxzz} \\ &+ \chi^{(3)}_{zzxz} + \chi^{(3)}_{zzzx}) + \sin^4\delta\cdot\chi^{(3)}_{zzzz}]. \end{aligned} \tag{28}$$

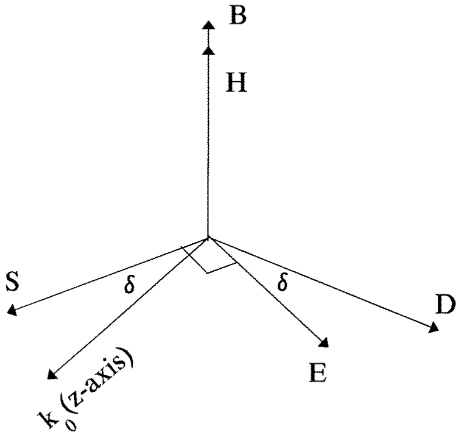

Figure 2. Direction of the field vectors, the Poynting vector $\boldsymbol{S}$ and the central wavevector of the beam $\boldsymbol{k}_0$ for the e-light beam propagating in uniaxial crystals. The electric field vector $\boldsymbol{E}$, the electric displacement vector $\boldsymbol{D}$, $\boldsymbol{S}$ and $\boldsymbol{k}_0$ are coplanar, and the magnetic field vector $\boldsymbol{H}$ (and hence also the magnetic induction vector $\boldsymbol{B}$) is at right angles to them. $\boldsymbol{D}$ is orthogonal to $\boldsymbol{k}_0$, but $\boldsymbol{E}$ is not. $\boldsymbol{S}$ is perpendicular to $\boldsymbol{E}$, but not to $\boldsymbol{D}$. The angle between $\boldsymbol{E}$ and $\boldsymbol{D}$ is the same as the angle between $\boldsymbol{S}$ and $\boldsymbol{k}_0$, and is denoted by δ.

Through the coordinate transformation shown in figure 1, n_2 can be expressed by the components in the principal coordinate system. For the $\bar{3}$m crystal, its n_2 is (details regarding the coordinate transformation are given in appendix B):

$$\begin{aligned} n_2^{(\bar{3}\mathrm{m})} = \frac{3}{8n^{\mathrm{e}}}\{&\cos^4\delta(\chi^{(3)\prime}_{x'x'x'x'}\cos^4\theta + \chi^{(3)\prime}_{z'z'z'z'}\sin^4\theta \\ &+\chi_1'\sin^2\theta\cos^2\theta - \chi_2'\cos^3\theta\sin\theta) \\ &+\cos^3\delta\sin\delta\{\sin(2\theta)[2(\chi^{(3)\prime}_{x'x'x'x'}\cos^2\theta \\ &-\chi^{(3)\prime}_{z'z'z'z'}\sin^2\theta) + \chi_1'(\sin^2\theta - \cos^2\theta)] \\ &+\chi_2'\cos^2\theta(\cos^2\theta - 3\sin^2\theta)\} \\ &+\cos^2\delta\sin^2\delta\{[6(\chi^{(3)\prime}_{x'x'x'x'} + \chi^{(3)\prime}_{z'z'z'z'}) \\ &-4\chi_1']\sin^2\theta\cos^2\theta \\ &+\chi_1'(\cos^4\theta + \sin^4\theta) + 3\chi_2'\cos\theta\sin\theta(\cos^2\theta - \sin^2\theta)\} \\ &+\cos\delta\sin^3\delta\{\sin(2\theta)[2(\chi^{(3)\prime}_{x'x'x'x'}\sin^2\theta - \chi^{(3)\prime}_{z'z'z'z'}\cos^2\theta) \\ &+\chi_1'(\cos^2\theta - \sin^2\theta)] + \chi_2'\sin^2\theta(3\cos^2\theta - \sin^2\theta)\} \\ &+\sin^4\delta(\chi^{(3)\prime}_{x'x'x'x'}\sin^4\theta + \chi^{(3)\prime}_{z'z'z'z'}\cos^4\theta + \chi_1'\sin^2\theta\cos^2\theta \\ &+\chi_2'\sin^3\theta\cos\theta)\}, \end{aligned} \tag{29}$$

where

$$\chi_1' = \chi^{(3)\prime}_{x'x'z'z'} + \chi^{(3)\prime}_{z'z'x'x'} + \chi^{(3)\prime}_{z'x'x'z'} + \chi^{(3)\prime}_{x'z'z'x'} + \chi^{(3)\prime}_{x'z'x'z'} + \chi^{(3)\prime}_{z'x'z'x'}, \tag{30}$$

and

$$\chi_2' = \chi^{(3)\prime}_{x'x'x'z'} + \chi^{(3)\prime}_{x'x'z'x'} + \chi^{(3)\prime}_{x'z'x'x'} + \chi^{(3)\prime}_{z'x'x'x'}. \tag{31}$$

The crystals 6/mmm and 4/mmm have the same nonzero elements, which are less than those of the crystal $\bar{3}$m. Therefore, their n_2 have a simpler expression:

$$\begin{aligned} n_2^{(6/\mathrm{mmm})} &= n_2^{(4/\mathrm{mmm})} \\ &= \frac{3}{8n^{\mathrm{e}}}\{\cos^4\delta(\chi^{(3)\prime}_{x'x'x'x'}\cos^4\theta + \chi^{(3)\prime}_{z'z'z'z'}\sin^4\theta \\ &\quad+\chi_1'\sin^2\theta\cos^2\theta) \\ &\quad+\cos^3\delta\sin\delta\sin(2\theta)[2(\chi^{(3)\prime}_{x'x'x'x'}\cos^2\theta \\ &\quad-\chi^{(3)\prime}_{z'z'z'z'}\sin^2\theta) + \chi_1'(\sin^2\theta - \cos^2\theta)] \\ &\quad+\cos^2\delta\sin^2\delta\{[6(\chi^{(3)\prime}_{x'x'x'x'} + \chi^{(3)\prime}_{z'z'z'z'}) \\ &\quad-4\chi_1']\sin^2\theta\cos^2\theta + \chi_1'(\cos^4\theta + \sin^4\theta)\} \\ &\quad+\cos\delta\sin^3\delta\sin(2\theta)[2(\chi^{(3)\prime}_{x'x'x'x'}\sin^2\theta \\ &\quad-\chi^{(3)\prime}_{z'z'z'z'}\cos^2\theta) + \chi_1'(\cos^2\theta - \sin^2\theta)] \\ &\quad+\sin^4\delta(\chi^{(3)\prime}_{x'x'x'x'}\sin^4\theta + \chi^{(3)\prime}_{z'z'z'z'}\cos^4\theta \\ &\quad+\chi_1'\sin^2\theta\cos^2\theta)\}. \end{aligned} \tag{32}$$

It is obvious that n_2 is anisotropic and dependent on the propagation direction. For example, when θ approaches zero (or π) or equals $\pi/2$ we obtain

$$\left.\begin{aligned} \lim_{\theta\to 0} \quad n_2 &= \\ \lim_{\theta\to \pi} \quad n_2 &= \end{aligned}\right\} n_2^{(\mathrm{o})} = \frac{3\chi^{(3)\prime}_{x'x'x'x'}}{8n_{\mathrm{o}}}, \qquad n_2\left(\frac{\pi}{2}\right) = \frac{3\chi^{(3)\prime}_{z'z'z'z'}}{8n_{\mathrm{e}}}, \tag{33}$$

for all three crystals, where $n_2^{(\mathrm{o})}$ is the NRI for the o-light.

Since the condition that

$$\Delta_L = \frac{(n_{\mathrm{o}} - n_{\mathrm{e}})}{n_{\mathrm{o}}} \ll 1 \tag{34}$$

is satisfied for most of uniaxial crystals [13], n_2 can be simplified greatly if its expression is expanded about small parameter Δ_L to the first order. First of all, n^{e} and δ can be expanded as

$$n^{\mathrm{e}} = n_{\mathrm{o}}(1 - \Delta_L\sin^2\theta) + \mathrm{o}(\Delta_L), \tag{35}$$

and

$$\delta = -\Delta_L\sin(2\theta) + \mathrm{o}(\Delta_L). \tag{36}$$

Then, equation (29) is reduced to

$$\begin{aligned} n_2^{(\bar{3}\mathrm{m})} = \frac{3}{8n_{\mathrm{o}}}\{&(1 + \Delta_L\sin^2\theta)[\chi^{(3)\prime}_{x'x'x'x'}\cos^4\theta \\ &+\chi^{(3)\prime}_{z'z'z'z'}\sin^4\theta + \chi_1'\sin^2\theta\cos^2\theta - \chi_2'\cos^3\theta\sin\theta] \\ &-\Delta_L\sin(2\theta)\{\sin(2\theta)[2(\chi^{(3)\prime}_{x'x'x'x'}\cos^2\theta - \chi^{(3)\prime}_{z'z'z'z'}\sin^2\theta) \\ &+\chi_1'(\sin^2\theta - \cos^2\theta)] + \chi_2'\cos^2\theta(\cos^2\theta - 3\sin^2\theta)\}\} \\ &+\mathrm{o}(\Delta_L), \end{aligned} \tag{37}$$

and the simplified expression of n_2 for crystals 6/mmm and 4/mmm is

$$\begin{aligned} n_2^{(6/\mathrm{mmm})} &= n_2^{(4/\mathrm{mmm})} \\ &= \frac{3}{8n_{\mathrm{o}}}\{(1 + \Delta_L\sin^2\theta)[\chi^{(3)\prime}_{x'x'x'x'}\cos^4\theta \\ &\quad+\chi^{(3)\prime}_{z'z'z'z'}\sin^4\theta + \chi_1'\sin^2\theta\cos^2\theta] \\ &\quad-\Delta_L\sin^2(2\theta)[2(\chi^{(3)\prime}_{x'x'x'x'}\cos^2\theta \\ &\quad-\chi^{(3)\prime}_{z'z'z'z'}\sin^2\theta) + \chi_1'(\sin^2\theta - \cos^2\theta)]\} \\ &\quad+\mathrm{o}(\Delta_L). \end{aligned} \tag{38}$$

Expressions (37) and (38) can still be further simplified by considering the Kleinman symmetry condition [14]. In this case, we can obtain

$$n_2^{(\bar{3}\mathrm{m})} = \frac{3}{8n_\mathrm{o}}\{(1+\Delta_L\sin^2\theta)[\chi^{(3)'}_{x'x'x'x'}\cos^4\theta + \chi^{(3)'}_{z'z'z'z'}\sin^4\theta + 6\chi^{(3)'}_{x'x'z'z'}\sin^2\theta\cos^2\theta - 4\chi^{(3)'}_{x'x'x'z'}\cos^3\sin\theta] - \Delta_L\sin(2\theta)\{\sin(2\theta)[2(\chi^{(3)'}_{x'x'x'x'}\cos^2\theta - \chi^{(3)'}_{z'z'z'z'}\sin^2\theta) + 6\chi^{(3)'}_{x'x'z'z'}(\sin^2\theta-\cos^2\theta)] + 4\chi^{(3)'}_{x'x'x'z'}\cos^2\theta(\cos^2\theta - 3\sin^2\theta)\}\} + \mathrm{o}(\Delta_L), \tag{39}$$

and

$$n_2^{(6/\mathrm{mmm})} = \frac{3}{8n_\mathrm{o}}\{(1+\Delta_L\sin^2\theta)[\chi^{(3)'}_{x'x'x'x'}\cos^4\theta + \chi^{(3)'}_{z'z'z'z'}\sin^4\theta + 6\chi^{(3)'}_{x'x'z'z'}\sin^2\theta\cos^2\theta] - \Delta_L\sin^2(2\theta)[2(\chi^{(3)'}_{x'x'x'x'}\cos^2\theta - \chi^{(3)'}_{z'z'z'z'}\sin^2\theta) + 6\chi^{(3)'}_{x'x'z'z'}(\sin^2\theta-\cos^2\theta)]\} + \mathrm{o}(\Delta_L). \tag{40}$$

4. Properties of the self-focusing and self-trapping of a light beam in uniaxial crystals

After obtaining the NRI, we are now in a position to discuss the behaviour of the paraxial beam in the uniaxial crystals when the diffraction and nonlinearity act together.

The introduction of equation (27) into (13) gives

$$\alpha_1 = \cos^2\delta = 1 + \mathrm{o}(\Delta_L^2) \approx 1, \tag{41}$$

and, hence, by combining with equation (24), wave equation (12) becomes

$$\mathrm{i}\left(\frac{\partial A}{\partial z} - \sigma\frac{\partial A}{\partial x}\right) + \frac{1}{2k_0}\left(\rho_x\frac{\partial^2 A}{\partial x^2} + \rho_y\frac{\partial^2 A}{\partial y^2}\right) + \frac{\omega n_2}{c}|A|^2 A = 0, \tag{42}$$

where

$$\sigma = (\nabla_K k_0)_{x'} \qquad \rho_x = -k_0(\nabla_K\nabla_K k_0)_{xx'} \qquad \rho_y = -k_0(\nabla_K\nabla_K k_0)_{yy} \tag{43}$$

for $0 < \theta < \pi$. If the nonlinear term is not considered, and the $[(\nabla_K k_0)_x]^2$ term in ρ_x which can be proved to be of the order of Δ_L^2, is dropped, the linear case of equation (42) is equal to that obtained by Fleck and Feit (equation (39) in [18]). However, our treatment is simpler.

Using the transformation

$$u(\xi,\eta,\zeta) = \sqrt{\frac{\omega n_2}{ck_0}}A(x,y,z), \tag{44a}$$

$$\xi = \frac{k_0(\sigma z + x)}{\sqrt{\rho_x}}, \tag{44b}$$

$$\eta = \frac{k_0 y}{\sqrt{\rho_y}}, \tag{44c}$$

$$\zeta = k_0 z, \tag{44d}$$

the transverse asymmetrical equation (42) is readily transformed to the transverse symmetric nonlinear Schrödinger equation:

$$\mathrm{i}\frac{\partial u}{\partial\zeta} + \frac{1}{2}\left(\frac{\partial^2 u}{\partial\xi^2} + \frac{\partial^2 u}{\partial\eta^2}\right) + |u|^2 u = 0. \tag{45}$$

Transformation (44b) shows that the propagation of the e-light beam is inclined to its central wavevector $k_0\boldsymbol{e}_z$. This deflection results from the fact that the e-light beam propagates along the direction of the Poynting vector rather than along the central wavevector $k_0\boldsymbol{e}_z$ [12, 19]. It is easy to see that the deflection equals zero in the direction perpendicular to the c-axis.

The analytical approaches to solving equation (45) are the aberrationless ray approximation [5,8] and the variational method [9]. The former provides some qualitatively correct answers but the quantitative predictions are less reliable, while the latter can give analytical results in good agreement with numerical results. Moreover, a sech trial function yields more accurate approximation than a Gaussian trial function [9], because the sech function is the exact profile for a (1 + 1)-dimensional nonlinear Schrödinger equation. Therefore, the variational approach with the sech ansatz is used here to find the solution of equation (45). For the choice of symmetry sech trial function

$$u(\xi,\eta,\zeta) = \Lambda(\zeta)\mathrm{sech}\left[\frac{\sqrt{\xi^2+\eta^2}}{a(\zeta)}\right]\exp[\mathrm{i}b(\zeta)(\xi^2+\eta^2)], \tag{46}$$

proves that if the beam waist is located at the entrance face of the media, that is $\mathrm{d}a(\zeta)/\mathrm{d}\zeta = 0$ at $\zeta = 0$, the normalized beam radius $a(\zeta)$ has the form

$$\frac{a(\zeta)^2}{a_0^2} = \frac{\kappa_1\zeta^2(1-\kappa_2|\Lambda_0|^2 a_0^2)}{a_0^4} + 1, \tag{47}$$

where a_0 and $|\Lambda_0|$ are the initial normalized beam radius and amplitude, respectively, and $\kappa_1 = 4(2\ln 2 + 1)/[27f(3)]$, $\kappa_2 = (4\ln 2 - 1)/(2\ln 2 + 1)$ ($f(n)$ denotes the Riemann zeta function, $f(3) \approx 1.202$). By transformation (44), we can get the real physical quantities

$$|A(x,y,z)| = \sqrt{\frac{ck_0}{\omega n_2}}|\Lambda(z)|\mathrm{sech}\left(\sqrt{\frac{(\sigma z + x)^2}{w_x^2(z)} + \frac{y^2}{w_y^2(z)}}\right), \tag{48}$$

where

$$W_x = \frac{a\sqrt{\rho_x}}{k_0}, \qquad W_y = \frac{a\sqrt{\rho_y}}{k_0} \tag{49}$$

are beam widths in the x-direction and y-direction, respectively, and

$$\frac{S(z)}{S_0} = \frac{\pi^2\kappa_1\rho_x\rho_y z^2}{S_0^2 k_0^2}\left(1 - \frac{P_0}{P_\mathrm{cre}}\right) + 1, \tag{50}$$

$$P_\mathrm{cre}(\theta) = P_\mathrm{cro}\sqrt{\rho_x\rho_y}\frac{n_2^{(\mathrm{o})}}{n_2(\theta)}, \qquad P_\mathrm{cro} = \frac{\ln 2}{4\pi}\frac{\lambda_0^2}{\kappa_2 Z_0 n_2^{(0)}}, \tag{51}$$

where $S = \pi w_x w_y$ is a beam area, S_0 its initial value, P_0 represents the input power [$P = (\frac{1}{2})\int \boldsymbol{E}\times\boldsymbol{H}^*\,\mathrm{d}x\mathrm{d}y = k_0/(2\mu_0\omega)\int|E|^2\,\mathrm{d}x\mathrm{d}y$], P_cre and P_cro are critical powers for the e-light and the o-light, respectively, $Z_0[= (\mu_0/\varepsilon_0)^{1/2} \approx 377\ \Omega]$ the vacuum impedance, and λ_0 is the wavelength in vacuum.

4.1. Critical power

Equation (50) tells us that when $P_0 = P_{\rm cre}$, the beam area keeps constant S_0. This is the self-trapping situation. If $P_0 > P_{\rm cre}$, S will reduce continuously to a point until the beam collapses, which is the self-focusing situation; on the contrary, the beam will diffract.

Substitution of the expressions for ρ_x, ρ_y and $n_2^{(0)}$ into equation (51) and by means of equation (39), $P_{\rm cre}$ of crystal $\bar{3}$m reads

$$\frac{P_{\rm cre}^{(\bar{3}{\rm m})}(\theta)}{P_{\rm cro}} = \frac{1 + 2\Delta_L(1-2\sin^2\theta) + {\rm o}(\Delta_L)}{D}, \qquad (52a)$$

where

$$D = 1 - \Delta_{N1}\sin^4\theta - \tfrac{1}{2}\Delta_{N2}\sin^2(2\theta) - 4\Delta_{N3}\cos^3\theta\sin\theta - \Delta_L\sin(2\theta)\{2\sin(2\theta)[\Delta_{N1}\sin^2\theta + \Delta_{N2}(1-2\sin^2\theta)] + 4\Delta_{N3}\cos^2\theta(1-4\sin^2\theta)\}, \qquad (52b)$$

$$\Delta_{N1} = \frac{\chi^{(3)'}_{x'x'x'x'} - \chi^{(3)'}_{z'z'z'z'}}{\chi^{(3)'}_{x'x'x'x'}},$$
$$\Delta_{N2} = \frac{\chi^{(3)'}_{x'x'x'x'} - 3\chi^{(3)'}_{x'x'z'z'}}{\chi^{(3)'}_{x'x'x'x'}}, \qquad \Delta_{N3} = \frac{\chi^{(3)'}_{x'x'x'z'}}{\chi^{(3)'}_{x'x'x'x'}}. \qquad (52c)$$

If Δ_{N1}, Δ_{N2} and Δ_{N3} are assumed to be small parameters ($\ll$1), expression (52a) can be reduced further as

$$\frac{P_{\rm cre}^{(\bar{3}{\rm m})}(\theta)}{P_{\rm cro}} = 1 + 2\Delta_L(1-2\sin^2\theta) + \Delta_{N1}\sin^4\theta + \tfrac{1}{2}\Delta_{N2}\sin^2(2\theta) + 4\Delta_{N3}\cos^3\theta\sin\theta + {\rm o}(\Delta_L + \Delta_{N1} + \Delta_{N2} + \Delta_{N3}). \qquad (53)$$

From equation (53), let $\Delta_{N3} = 0$, i.e. $\chi^{(3)'}_{x'x'x'z'} = 0$, we can obtain the critical power for crystal 6/mmm (and 4/mmm):

$$\frac{P_{\rm cre}^{(6/{\rm mmm})}(\theta)}{P_{\rm cro}} = 1 + 2\Delta_L(1-2\sin^2\theta) + \Delta_{N1}\sin^4\theta + \tfrac{1}{2}\Delta_{N2}\sin^2(2\theta) + {\rm o}(\Delta_L + \Delta_{N1} + \Delta_{N2}), \qquad (54)$$

where both Δ_{N1} and Δ_{N2} are also assumed to be small parameters.

We can conclude from equations (51), (53) and (54) that the e-light beam propagating in a different direction has a different critical power, but the o-light's critical power is independent of the propagation direction, as in the case of isotropic media.

Figures 3 and 4 pertain to the critical power as the function of θ obtained from equation (53) for the case where $\Delta_L \approx 0.158$. In the figures, Δ_{N1}, Δ_{N2} and Δ_{N3} are all, as assumed, small parameters, their absolute values being less than 0.2, because no such data are available both experimentally and theoretically. These figures correspond to the critical power of negative crystal $NaNO_3$, whose crystal class is $\bar{3}$m [14], and where $n_{\rm o}$ and $n_{\rm e}$ are equal to 1.587 and 1.336, respectively [13]. The curves start from the same point $(1+2\Delta_L)$ at $\theta = 0$, reach a extreme $(1-2\Delta_L+\Delta_{N1})$ at $\theta = \pi/2$, which is the minimum for some of the three parameters Δ_{N1}, Δ_{N2} and Δ_{N3}, and end in the same point $(1+2\Delta_L)$ at $\theta = \pi$. The figures also show that $P_{\rm cre}$ might have the other extremes depending upon the value of the parameters Δ_{N1}, Δ_{N2} and Δ_{N3}. We find from the figures that the minimum of the difference between the maximum and the minimum of the critical power $((|P_{\rm cre}/P_{\rm cro})_{\max} - (P_{\rm cre}/P_{\rm cro})_{\min})$ is about

$$\left.\frac{P_{\rm cre}(0) - P_{\rm cre}(\pi/2)}{P_0}\right|_{\Delta_{N1}=0.2,\Delta_{N3}=0.0,\ \text{any}\ \Delta_{N2}} = (4\Delta_L - \Delta_{N1})|_{\Delta_L=0.158,\Delta_{N1}=0.2} \approx 43\%. \qquad (55)$$

This means that no matter how many unknown parameters Δ_{N1}, Δ_{N2} and Δ_{N3} there are, the difference $(|P_{\rm cre}/P_{\rm cro})_{\max} - (P_{\rm cre}/P_{\rm cro})_{\min}$ in the crystal $\bar{3}$m is large enough to be easily detected experimentally. Therefore, the measurements of critical power at various directions relative to the optic axis of the crystal make it possible to obtain a set of independent equations from equation (53) to calculate the independent nonzero components of its $\chi^{(3)}$ tensor.

The analytical analysis about the expression of critical power becomes possible for the crystal 6/mmm (and also 4/mmm) because its Δ_{N3} is zero. The analysis of equation (54) shows that the critical power has two extremes: $P_{\rm cre} \to P_{\rm ext1} = (1+2\Delta_L)P_{\rm cro}$ when θ goes to 0 (or π), and $P_{\rm cre}$ equals $P_{\rm ext2} = (1-2\Delta_L+\Delta_{N1})P_{\rm cro}$ at $\theta = \pi/2$. There is another extreme:

$$P_{\rm ext3} = \frac{\Delta_{N1} + 2\Delta_L(\Delta_{N1} - 2\Delta_L) - \Delta_{N2}(2+\Delta_{N2})}{\Delta_{N1} - 2\Delta_{N2}} \qquad (56)$$

which at θ equals $\theta_c = \arcsin[(2\Delta_L - \Delta_{N2})/(\Delta_{N1} - 2\Delta_{N2})]^{1/2}$, if a condition $0 < (2\Delta_L - \Delta_{N2})/(\Delta_{N1} - 2\Delta_{N2}) \leqslant 1$ is satisfied. Figure 5 gives the critical power as a function of θ for a positive crystal Rutile (its crystal class is 4/mmm [14], and its $n_{\rm o} = 2.616$, and $n_{\rm e} = 2.903$ [13], so that $\Delta_L \approx -0.110$), where both Δ_{N1} and Δ_{N2} are also small parameters, and the absolute value is less than 0.2. For the case where $\Delta_{N1} = 0.2$ and any Δ_{N2}, and the case where $\Delta_{N1} = 0.1$ and any Δ_{N2}, $P_{\rm cre}$ is single-valued function of θ in the interval $(0, \pi/2)$ because the condition that θ_c appears can never be satisfied. However, when Δ_{N1} is small enough, the function $P_{\rm cre}(\theta)$ becomes a little complex and, as shown in the figure, the third extreme as a maximum might appear or not, depending upon the value of Δ_{N2}. Figure 5 shows that the difference between the maximum and the minimum of $P_{\rm cre}$ is at least about

$$\left.\frac{P_{\rm cre}(\pi/2) - P_{\rm cre}(0)}{P_0}\right|_{\Delta_{N1}=\Delta_{N2}=-0.2} = (-4\Delta_L + \Delta_{N1})|_{\Delta_L=-0.11,\Delta_{N1}=-0.2} = 24\%, \qquad (57)$$

when $\Delta_{N1} = -0.2$, or bigger if Δ_{N1} is bigger, which should be easily measurable.

4.2. Self-focusing length

A self-focusing length is defined as the distance where the beam spot shrinks to zero and the optical beam collapses. From equation (50), letting $S = 0$ we can obtain the self-focusing length of the e-light, which reads

$$z_f = \frac{S_0 k_0}{\pi\kappa_1^{\frac{1}{2}}} \frac{\beta_1(\theta)}{\sqrt{\frac{P_0}{P_{\rm cro}}\beta_2(\theta) - 1}} \qquad (58)$$

where

$$\beta_1(\theta) = \frac{1}{\sqrt{\rho_x\rho_y}} = 1 + \Delta_L(1 - 3\cos^2\theta) + {\rm o}(\Delta_L), \qquad (59)$$

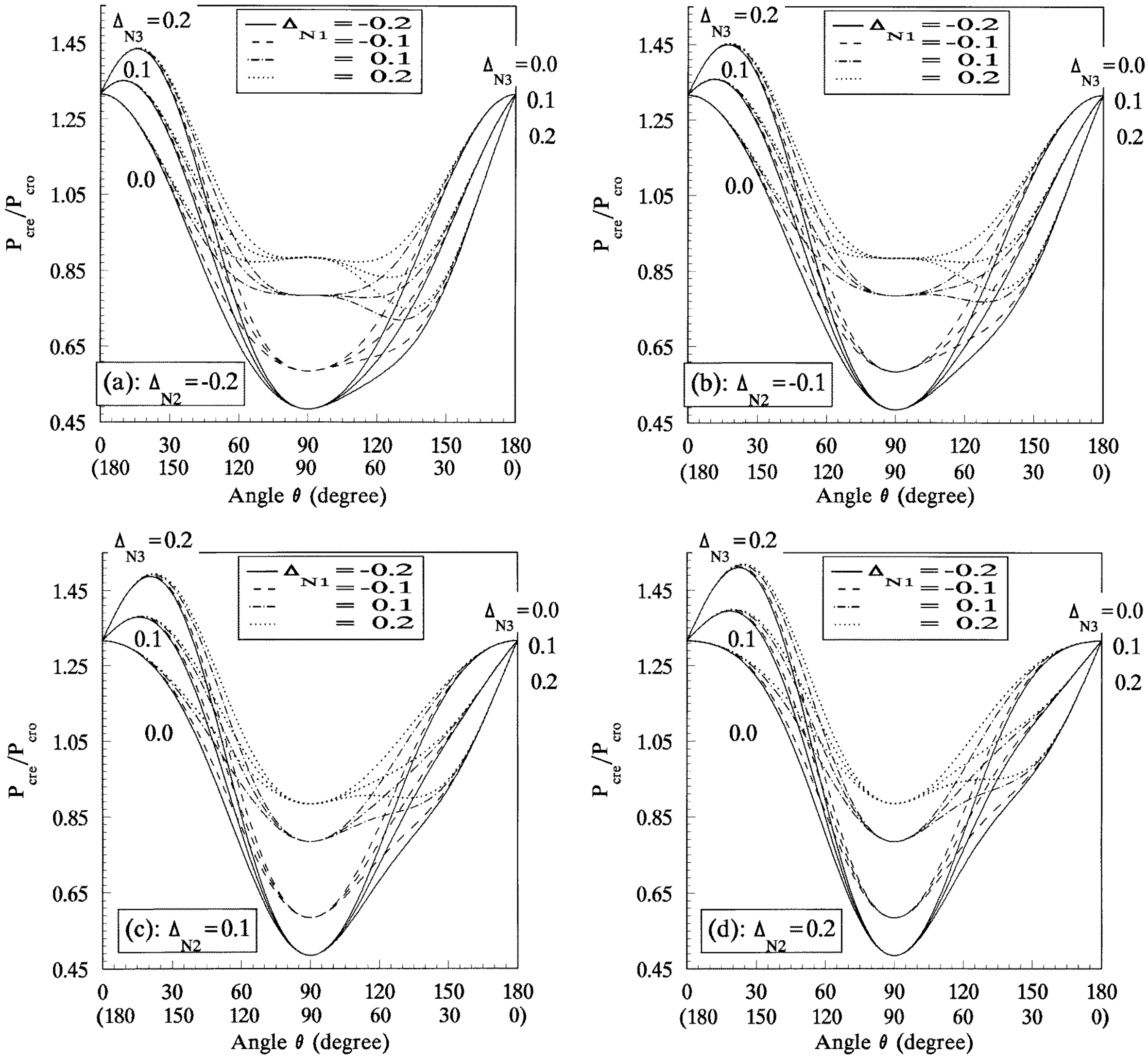

Figure 3. Critical powers as the function of θ for the negative crystal $NaNO_3$, where Δ_{N1}, Δ_{N2} and Δ_{N3} are all assumed as small parameters, with their absolute values less than 0.2. The θ coordinate values in the first row are for the Δ_{N3} given in the figures, and the ones in the second row are for the minus of the Δ_{N3} given, because equation (53) is unchangeable through a transformation such that $\theta = \pi - \theta$ and $\Delta_{N3} = -\Delta_{N3}$.

and

$$\beta_2(\theta) = \frac{n_2(\theta)/n_2^{(o)}}{\sqrt{\rho_x\rho_y}}$$
$$= \begin{cases} 1 - 2\Delta_L(1-2\sin^2\theta) - \Delta_{N1}\sin^4\theta - \frac{1}{2}\Delta_{N2}\sin^2(2\theta) \\ -4\Delta_{N3}\cos^3\theta\sin\theta \\ +\mathrm{o}(\Delta_L+\Delta_{N1}+\Delta_{N2}+\Delta_{N3}), \quad \text{for crystal } \bar{3}\text{m}; \\ 1 - 2\Delta_L(1-2\sin^2\theta) - \Delta_{N1}\sin^4\theta - \frac{1}{2}\Delta_{N2}\sin^2(2\theta) \\ +\mathrm{o}(\Delta_L+\Delta_{N1}+\Delta_{N2}), \quad \text{for crystal 6/mmm.} \end{cases} \tag{60}$$

Equation (58) will give the self-focusing length of the o-light if β_1 and β_2 are set to be 1, letting $\Delta_L = \Delta_{N1} = \Delta_{N2} = \Delta_{N3} = 0$ in equations (59) and (60). It is obvious that the self-focusing length is different for the e-light at a different direction, but isotropic for the o-light.

As has been pointed out [20,21], however, this collapse is due to the loss of validity of the nonlinear paraxial wave equation (12) in the neighbourhood of the self-focus. To deal with the detail in the neighbourhood of the self-focus in the crystals, the other models should be used as in the case of isotropic media [20,21].

4.3. Light spot of the self-trapping beam

We can demonstrate that the self-trapping beam of the e-light is asymmetrical, and its light spot is an ellipse. The ratio of the two axes of this ellipse (which axis is the major axis depends on the sign of Δ_L) equals

$$\frac{W_x}{W_y} = \sqrt{\frac{\rho_x}{\rho_y}} = 1 - \Delta_L\sin^2\theta + \mathrm{o}(\Delta_L). \tag{61}$$

The result tells us that the asymmetry becomes strongest at $\theta = \pi/2$, and the ellipse becomes a circle as θ goes to zero or π. It goes without saying that the self-trapping beam for the o-light is circular, as that in the isotropic media.

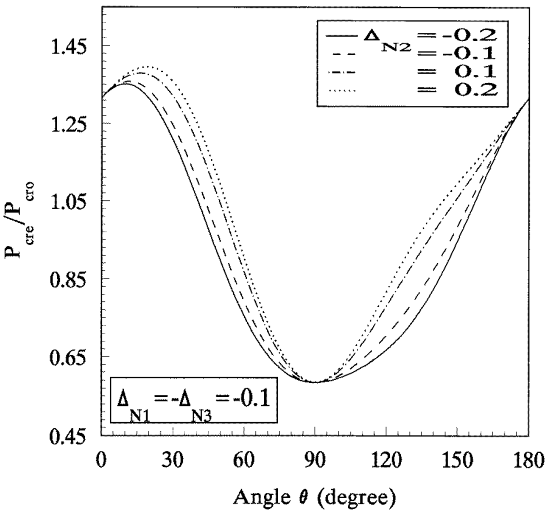

Figure 4. Critical powers as the function of θ for the negative crystal $NaNO_3$, where $\Delta_{N1} = -\Delta_{N3} = -0.1$, and Δ_{N2} is as the small parameter; its absolute values less than 0.2.

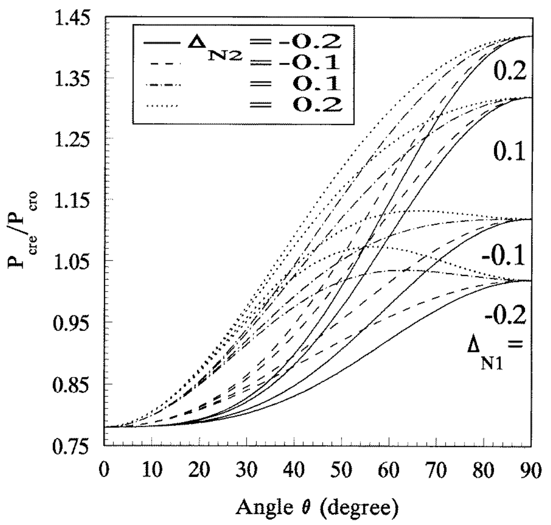

Figure 5. Critical powers as the function of θ for the positive crystal Rutile, where Δ_{N1} and Δ_{N2} are as parameters, with their absolute values less than 0.2. Because the curves are symmetrical about $\theta = \pi/2$ in the interval $(0, \pi)$, just half of them are drawn.

5. Conclusion

We have derived the nonlinear paraxial wave equation describing the propagation of the optical beam in nonlinear anisotropic media with centrosymmetry. As for application, we obtained the nonlinear refractive index in the three uniaxial crystals belonging to the symmetry classes 6/mmm in the hexagonal system, 4/mmm in the tetragonal system and $\bar{3}$m in the trigonal system, respectively, and considered the self-trapping and self-focusing of the optical beam propagating along any direction in these crystals. The nonlinear refractive index, critical power and self-focusing length are all anisotropic for the e-light, that is, they are dependent on the propagation direction. However, these quantities are isotropic for the o-light. There exists an elliptical self-trapping beam for the e-light. These features can be easily understood considering the anisotropy for the e-light and isotropy for the o-light. Our results might be applied to the experimental determination of the anisotropy of $\chi^{(3)}$.

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Appendix A. Proof of neglecting the terms that come from $\nabla \cdot \boldsymbol{E}$

By use of expansion (4*b*) and one of the properties of the Fourier transform, which reads

$$\iint \boldsymbol{K}\boldsymbol{E}_0^{(0)} \exp[\mathrm{i}\varphi(\boldsymbol{K}, \boldsymbol{R}, z)]\,\mathrm{d}\boldsymbol{K} = -\mathrm{i}\nabla_\perp A\boldsymbol{e}^0,$$

the first two terms in equation (10) can be reduced as

$$\begin{aligned}
&\iint (k - k_0)(\boldsymbol{K}\boldsymbol{e}_z + \boldsymbol{e}_z\boldsymbol{K}) \cdot \boldsymbol{E}_0^{(0)} \exp(\mathrm{i}\varphi)\,\mathrm{d}\boldsymbol{K} \\
&\quad +(\nabla_\perp \boldsymbol{e}_z + \boldsymbol{e}_z\nabla_\perp) \cdot \boldsymbol{e}^0 \frac{\partial A}{\partial z} \\
&\approx \iint (\nabla_K k_0 \cdot \boldsymbol{K} + \tfrac{1}{2}\nabla_K\nabla_K k_0 : \boldsymbol{K}\boldsymbol{K}) \\
&\quad \times(\boldsymbol{K}\boldsymbol{e}_z + \boldsymbol{e}_z\boldsymbol{K}) \cdot \boldsymbol{E}_0^{(0)} \exp(\mathrm{i}\varphi)\,\mathrm{d}\boldsymbol{K} \\
&\quad +(\nabla_\perp \boldsymbol{e}_z + \boldsymbol{e}_z\nabla_\perp) \cdot \boldsymbol{e}^0 \frac{\partial A}{\partial z} \\
&= (\nabla_\perp \boldsymbol{e}_z + \boldsymbol{e}_z\nabla_\perp) \\
&\quad \cdot \boldsymbol{e}^0 \left(\frac{\partial A}{\partial z} - \nabla_K k_0 \cdot \nabla_\perp A + \mathrm{i}\frac{1}{2}\nabla_K\nabla_K k_0 : \nabla_\perp\nabla_\perp A\right).
\end{aligned} \tag{A.1}$$

By means of the order-of-magnitude analysis method [21], we can get the following relation of the order-of-magnitude:

$$\begin{aligned}
&\frac{\text{the first two terms in equation (11)}}{\text{the third term in equation (11)}} \\
&= \left\{\iint (k - k_0)(\boldsymbol{K}\boldsymbol{e}_z + \boldsymbol{e}_z\boldsymbol{K}) \cdot \boldsymbol{E}_0^{(0)} \exp(\mathrm{i}\varphi)\,\mathrm{d}\boldsymbol{K} \right.\\
&\quad \left. +(\nabla_\perp \boldsymbol{e}_z + \boldsymbol{e}_z\nabla_\perp) \cdot \boldsymbol{e}^0 \frac{\partial A}{\partial z}\right\}\left\{(\boldsymbol{e}_z\boldsymbol{e}_z - \hat{I}f)\right. \\
&\quad \left.\cdot\left[2\mathrm{i}k_0\boldsymbol{e}^0\frac{\partial A}{\partial z} + \boldsymbol{E}_0^{(0)}\iint (k^2 - k_0^2)\exp(\mathrm{i}\varphi)\,\mathrm{d}\boldsymbol{K}\right]\right\}^{-1} \\
&\sim \frac{\lambda_0}{2\pi w},
\end{aligned} \tag{A.2}$$

where w is the beam width. Generally, $\lambda_0/(2\pi w)$ is a very small parameter ($\ll 1$), which is just about 0.16 even if the beam is focused to $w \approx \lambda_0$. Therefore, the first two terms of equation (10) can be neglected in comparison with the other terms.

Furthermore, it can also be proved that the second derivative $\partial^2 A/\partial z^2$ is two orders of $\lambda_0/(2\pi w)$ less than the terms retained by the order-of-magnitude analysis method.

Appendix B. Calculation of the $\chi^{(3)}$ components under transformation from the principal coordinate system (x', y', z') to the propagation coordinate system (x, y, z)

For the $\bar{3}$m crystal, we can obtain the following from equation (17) and by means of equation (18):

$$\begin{aligned}\chi^{(3)}_{zxxx} &= \sum_{i',j',k',l'}^{(i',j',k',l'\neq y')} \chi^{(3)\prime}_{i'j'k'l'} g_{i'z} g_{j'x} g_{k'x} g_{l'x} \\ &= (\chi^{(3)\prime}_{x'x'x'x'} - \chi^{(3)\prime}_{z'z'x'x'} - \chi^{(3)\prime}_{z'x'x'z'} \\ &\quad - \chi^{(3)\prime}_{z'x'z'x'}) \cos^3\theta \sin\theta \\ &\quad + (-\chi^{(3)\prime}_{z'z'z'z'} + \chi^{(3)\prime}_{x'x'z'z'} + \chi^{(3)\prime}_{x'z'z'x'} \\ &\quad + \chi^{(3)\prime}_{x'z'x'z'}) \sin^3\theta \cos\theta \\ &\quad - (\chi^{(3)\prime}_{x'x'x'z'} + \chi^{(3)\prime}_{x'x'z'x'} + \chi^{(3)\prime}_{x'z'x'x'}) \sin^2\theta \cos^2\theta \\ &\quad + \chi^{(3)\prime}_{z'x'x'x'} \cos^4\theta. \end{aligned} \tag{B.1}$$

In the same way, we can get

$$\begin{aligned}\chi^{(3)}_{xzxx} &= (\chi^{(3)\prime}_{x'x'x'x'} - \chi^{(3)\prime}_{z'z'x'x'} - \chi^{(3)\prime}_{x'z'z'x'} \\ &\quad - \chi^{(3)\prime}_{x'z'x'z'}) \cos^3\theta \sin\theta \\ &\quad + (-\chi^{(3)\prime}_{z'z'z'z'} + \chi^{(3)\prime}_{x'x'z'z'} + \chi^{(3)\prime}_{z'x'x'z'} \\ &\quad + \chi^{(3)\prime}_{z'x'z'x'}) \sin^3\theta \cos\theta \\ &\quad - (\chi^{(3)\prime}_{x'x'x'z'} + \chi^{(3)\prime}_{x'x'z'x'} + \chi^{(3)\prime}_{z'x'x'x'}) \sin^2\theta \cos^2\theta \\ &\quad + \chi^{(3)\prime}_{x'z'x'x'} \cos^4\theta, \end{aligned} \tag{B.2}$$

$$\begin{aligned}\chi^{(3)}_{xxzx} &= (\chi^{(3)\prime}_{x'x'x'x'} - \chi^{(3)\prime}_{x'x'z'z'} - \chi^{(3)\prime}_{x'z'z'x'} \\ &\quad - \chi^{(3)\prime}_{z'x'z'x'}) \cos^3\theta \sin\theta \\ &\quad + (-\chi^{(3)\prime}_{z'z'z'z'} + \chi^{(3)\prime}_{z'z'x'x'} + \chi^{(3)\prime}_{z'x'x'z'} \\ &\quad + \chi^{(3)\prime}_{x'z'x'z'}) \sin^3\theta \cos\theta \\ &\quad - (\chi^{(3)\prime}_{x'x'x'z'} + \chi^{(3)\prime}_{x'z'x'x'} + \chi^{(3)\prime}_{z'x'x'x'}) \sin^2\theta \cos^2\theta \\ &\quad + \chi^{(3)\prime}_{x'x'z'x'} \cos^4\theta, \end{aligned} \tag{B.3}$$

and

$$\begin{aligned}\chi^{(3)}_{xxxz} &= (\chi^{(3)\prime}_{x'x'x'x'} - \chi^{(3)\prime}_{x'x'z'z'} - \chi^{(3)\prime}_{z'x'x'z'} \\ &\quad - \chi^{(3)\prime}_{x'z'x'z'}) \cos^3\theta \sin\theta \\ &\quad + (-\chi^{(3)\prime}_{z'z'z'z'} + \chi^{(3)\prime}_{z'z'x'x'} + \chi^{(3)\prime}_{x'z'z'x'} \\ &\quad + \chi^{(3)\prime}_{z'x'z'x'}) \sin^3\theta \cos\theta \\ &\quad - (\chi^{(3)\prime}_{z'x'x'x'} + \chi^{(3)\prime}_{x'z'x'x'} + \chi^{(3)\prime}_{x'x'z'x'}) \sin^2\theta \cos^2\theta \\ &\quad + \chi^{(3)\prime}_{x'x'x'z'} \cos^4\theta. \end{aligned} \tag{B.4}$$

The addition of equations (B.1)–(B.4) gives the coefficient of $\cos^3\delta \sin\delta$ in equation (29).

Similarly, we can also obtain

$$\begin{aligned}\chi^{(3)}_{zzxx} &= \chi^{(3)\prime}_{z'z'x'x'} \cos^4\theta + \chi^{(3)\prime}_{x'x'z'z'} \sin^4\theta + (\chi^{(3)\prime}_{x'x'x'x'} \\ &\quad + \chi^{(3)\prime}_{z'z'z'z'} - \chi^{(3)\prime}_{x'z'z'x'} - \chi^{(3)\prime}_{z'x'x'z'} \\ &\quad - \chi^{(3)\prime}_{x'z'x'z'} - \chi^{(3)\prime}_{z'x'z'x'}) \cos^2\theta \sin^2\theta \\ &\quad + (\chi^{(3)\prime}_{x'z'x'x'} + \chi^{(3)\prime}_{z'x'x'x'}) \cos^3\theta \sin\theta \\ &\quad - (\chi^{(3)\prime}_{x'x'x'z'} + \chi^{(3)\prime}_{x'x'z'x'}) \cos\theta \sin^3\theta, \end{aligned} \tag{B.5}$$

$$\begin{aligned}\chi^{(3)}_{zxxz} &= \chi^{(3)\prime}_{z'x'x'z'} \cos^4\theta + \chi^{(3)\prime}_{x'z'z'x'} \sin^4\theta \\ &\quad + (\chi^{(3)\prime}_{x'x'x'x'} + \chi^{(3)\prime}_{z'z'z'z'} - \chi^{(3)\prime}_{x'x'z'z'} - \chi^{(3)\prime}_{z'z'x'x'} \\ &\quad - \chi^{(3)\prime}_{x'z'x'z'} - \chi^{(3)\prime}_{z'x'z'x'}) \cos^2\theta \sin^2\theta \\ &\quad + (\chi^{(3)\prime}_{x'x'x'z'} + \chi^{(3)\prime}_{z'x'x'x'}) \cos^3\theta \sin\theta \\ &\quad - (\chi^{(3)\prime}_{x'x'z'x'} + \chi^{(3)\prime}_{x'z'x'x'}) \cos\theta \sin^3\theta, \end{aligned} \tag{B.6}$$

$$\begin{aligned}\chi^{(3)}_{zxzx} &= \chi^{(3)\prime}_{z'x'z'x'} \cos^4\theta + \chi^{(3)\prime}_{x'z'x'z'} \sin^4\theta \\ &\quad + (\chi^{(3)\prime}_{x'x'x'x'} + \chi^{(3)\prime}_{z'z'z'z'} - \chi^{(3)\prime}_{x'z'z'x'} - \chi^{(3)\prime}_{z'x'x'z'} \\ &\quad - \chi^{(3)\prime}_{x'x'z'z'} - \chi^{(3)\prime}_{z'z'x'x'}) \cos^2\theta \sin^2\theta \\ &\quad + (\chi^{(3)\prime}_{z'x'x'x'} + \chi^{(3)\prime}_{x'x'z'x'}) \cos^3\theta \sin\theta \\ &\quad - (\chi^{(3)\prime}_{x'z'x'x'} + \chi^{(3)\prime}_{x'x'x'z'}) \cos\theta \sin^3\theta, \end{aligned} \tag{B.7}$$

$$\begin{aligned}\chi^{(3)}_{xzzx} &= \chi^{(3)\prime}_{x'z'z'x'} \cos^4\theta + \chi^{(3)\prime}_{z'x'x'z'} \sin^4\theta \\ &\quad + (\chi^{(3)\prime}_{x'x'x'x'} + \chi^{(3)\prime}_{z'z'z'z'} - \chi^{(3)\prime}_{z'z'x'x'} - \chi^{(3)\prime}_{x'x'z'z'} \\ &\quad - \chi^{(3)\prime}_{x'z'x'z'} - \chi^{(3)\prime}_{z'x'z'x'}) \cos^2\theta \sin^2\theta \\ &\quad + (\chi^{(3)\prime}_{x'z'x'x'} + \chi^{(3)\prime}_{x'x'z'x'}) \cos^3\theta \sin\theta \\ &\quad - (\chi^{(3)\prime}_{x'x'x'z'} + \chi^{(3)\prime}_{z'x'x'x'}) \cos\theta \sin^3\theta, \end{aligned} \tag{B.8}$$

$$\begin{aligned}\chi^{(3)}_{xzxz} &= \chi^{(3)\prime}_{x'z'x'z'} \cos^4\theta + \chi^{(3)\prime}_{z'x'z'x'} \sin^4\theta \\ &\quad + (\chi^{(3)\prime}_{x'x'x'x'} + \chi^{(3)\prime}_{z'z'z'z'} - \chi^{(3)\prime}_{x'z'z'x'} - \chi^{(3)\prime}_{z'x'x'z'} \\ &\quad - \chi^{(3)\prime}_{x'x'z'z'} - \chi^{(3)\prime}_{z'z'x'x'}) \cos^2\theta \sin^2\theta \\ &\quad + (\chi^{(3)\prime}_{x'z'x'x'} + \chi^{(3)\prime}_{x'x'x'z'}) \cos^3\theta \sin\theta \\ &\quad - (\chi^{(3)\prime}_{z'x'x'x'} + \chi^{(3)\prime}_{x'x'z'x'}) \cos\theta \sin^3\theta, \end{aligned} \tag{B.9}$$

and

$$\begin{aligned}\chi^{(3)}_{xxzz} &= \chi^{(3)\prime}_{x'x'z'z'} \cos^4\theta + \chi^{(3)\prime}_{z'z'x'x'} \sin^4\theta \\ &\quad + (\chi^{(3)\prime}_{x'x'x'x'} + \chi^{(3)\prime}_{z'z'z'z'} - \chi^{(3)\prime}_{x'z'z'x'} - \chi^{(3)\prime}_{z'x'x'z'} \\ &\quad - \chi^{(3)\prime}_{x'z'x'z'} - \chi^{(3)\prime}_{z'x'z'x'}) \cos^2\theta \sin^2\theta \\ &\quad + (\chi^{(3)\prime}_{x'x'x'z'} + \chi^{(3)\prime}_{x'x'z'x'}) \cos^3\theta \sin\theta \\ &\quad - (\chi^{(3)\prime}_{z'x'x'x'} + \chi^{(3)\prime}_{x'z'x'x'}) \cos\theta \sin^3\theta, \end{aligned} \tag{B.10}$$

as well as

$$\begin{aligned}\chi^{(3)}_{xzzz} &= (\chi^{(3)\prime}_{x'x'x'x'} - \chi^{(3)\prime}_{z'z'x'x'} - \chi^{(3)\prime}_{z'x'x'z'} \\ &\quad - \chi^{(3)\prime}_{z'x'z'x'}) \cos\theta \sin^3\theta \\ &\quad + (-\chi^{(3)\prime}_{z'z'z'z'} + \chi^{(3)\prime}_{x'x'z'z'} + \chi^{(3)\prime}_{x'z'z'x'} \\ &\quad + \chi^{(3)\prime}_{x'z'x'z'}) \cos^3\theta \sin\theta \\ &\quad + (\chi^{(3)\prime}_{x'x'x'z'} + \chi^{(3)\prime}_{x'x'z'x'} + \chi^{(3)\prime}_{x'z'x'x'}) \cos^2\theta \sin^2\theta \\ &\quad - \chi^{(3)\prime}_{z'x'x'x'} \sin^4\theta, \end{aligned} \tag{B.11}$$

$$\begin{aligned}\chi^{(3)}_{zxzz} &= (\chi^{(3)\prime}_{x'x'x'x'} - \chi^{(3)\prime}_{z'z'x'x'} - \chi^{(3)\prime}_{x'z'z'x'} \\ &\quad - \chi^{(3)\prime}_{x'z'x'z'}) \cos\theta \sin^3\theta \\ &\quad + (-\chi^{(3)\prime}_{z'z'z'z'} + \chi^{(3)\prime}_{x'x'z'z'} + \chi^{(3)\prime}_{z'x'x'z'} \\ &\quad + \chi^{(3)\prime}_{z'x'z'x'}) \cos^3\theta \sin\theta \\ &\quad + (\chi^{(3)\prime}_{x'x'x'z'} + \chi^{(3)\prime}_{x'x'z'x'} + \chi^{(3)\prime}_{z'x'x'x'}) \cos^2\theta \sin^2\theta \\ &\quad - \chi^{(3)\prime}_{x'z'x'x'} \sin^4\theta, \end{aligned} \tag{B.12}$$

$$\begin{aligned}\chi^{(3)}_{zzxz} &= (\chi^{(3)\prime}_{x'x'x'x'} - \chi^{(3)\prime}_{x'x'z'z'} - \chi^{(3)\prime}_{x'z'z'x'} \\ &\quad - \chi^{(3)\prime}_{z'x'z'x'}) \cos\theta \sin^3\theta \\ &\quad + (-\chi^{(3)\prime}_{z'z'z'z'} + \chi^{(3)\prime}_{z'z'x'x'} + \chi^{(3)\prime}_{z'x'x'z'} + \chi^{(3)\prime}_{x'z'x'z'}) \cos^3\theta \sin\theta \\ &\quad + (\chi^{(3)\prime}_{x'x'x'z'} + \chi^{(3)\prime}_{x'z'x'x'} + \chi^{(3)\prime}_{z'x'x'x'}) \cos^2\theta \sin^2\theta \\ &\quad - \chi^{(3)\prime}_{x'x'z'x'} \sin^4\theta, \end{aligned} \tag{B.13}$$

and

$$\chi^{(3)}_{zzzx} = (\chi^{(3)\prime}_{x'x'x'x'} - \chi^{(3)\prime}_{x'x'z'z'} - \chi^{(3)\prime}_{z'x'x'z'} - \chi^{(3)\prime}_{x'z'x'z'}) \cos\theta \sin^3\theta$$
$$+(-\chi^{(3)\prime}_{z'z'z'z'} + \chi^{(3)\prime}_{z'z'x'x'} + \chi^{(3)\prime}_{x'z'z'x'}$$
$$+\chi^{(3)\prime}_{z'x'z'x'}) \cos^3\theta \sin\theta$$
$$+(\chi^{(3)\prime}_{x'x'z'x'} + \chi^{(3)\prime}_{x'z'x'x'} + \chi^{(3)\prime}_{z'x'x'x'}) \cos^2\theta \sin^2\theta$$
$$-\chi^{(3)\prime}_{x'x'x'z'} \sin^4\theta. \quad \text{(B.14)}$$

Addition from equation (B.5) to equation (B.10) and from equation (B.11) to equation (B.14) can obtain the two corresponding expressions in equation (29), respectively.

Finally, the transformed results of $\chi^{(3)}_{xxxx}$ and $\chi^{(3)}_{zzzz}$ can be obtained directly from equations (17) and (18), which can be found in equation (29).

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