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Chirality versus dichroism: competition and role in conical diffraction displayed from the Nd:Bi₂ZnOB₂O₆ acentric biaxial laser crystal

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Abstract : We have grown acentric chiral Nd-doped BZBO single crystals by the Kyropoulos method, cut perpendicular to the optical axis. With a focused beam at wavelengths in the spectral domain of transparency with various polarizations, the conoscopy patterns revealed the specific role of chirality: disappearance of the optical axis replaced by two C-directions, linear polarization rotation and spiral patterns. The specific rotatory power was measured. The output near field exhibits the influence of chirality on the conical diffraction. Going to wavelengths in the spectral domain of absorption corresponding to the $^4I_{9/2} \rightarrow ^4F_{5/2}$ Nd³⁺ transition, we first studied the chirality/dichroism competition from propagation along the C-directions, characterizing the circular and elliptic eigen-modes. We exhibited the wavelengths at which the ellipse degenerates linearly, signature of the chirality/dichroism balancing. The second aspect is the existence of the optical axis which can be restored if the dichroism is high enough. In this case a singular Voigt wave can propagate. From the experimental determination of the optical parameters, a theoretical description has been proposed, leading to quite well agreement in the far field (conoscopy) and in the near field (conical diffraction).

1. Introduction

Acentric borate crystals have received much attention because their quadratic nonlinearities are phase-matchable for frequency conversion processes reaching the UV spectral range. Simultaneously, they have a high transparency in this domain with a high optical damage threshold. Among the most known crystals [1], we can cite YAl₃(BO₃)₄, ²-BaB₂O₄, LiB₃O₅, CsB₃O₅, CsLiB₆O₁₀, La₂CaB₁₀O₁₉ or BiB₃O₆.

Recently a novel acentric oxy-borate crystal, Bi₂ZnOB₂O₆ (BZBO), has been elaborated in bulk size by Kyropoulos technique [2] and top-seeded growth method [3]. The crystal was found non-hygroscopic and stable in a moist environment. Infra-red spectroscopy, transmittance and refractive indices measurements have been carried out. It was established to be phase matchable for second harmonic generation of the 1064 nm YAG:Nd laser radiation and its nonlinear optical coefficients have been measured by the Maker fringes method.

Despite that BZBO is a biaxial crystal, the light propagation along an optical axis was not investigated up to now. This propagation direction is of particular interest because several spectacular and intriguing optical phenomena occur. A focused flux of light energy is split into a cone inside the crystal and finally rings come out instead of the double-refraction observed in the other wave directions: it is the internal conical refraction/diffraction [4-5] predicted in 1832 by W. Hamilton. In the early times it was studied by a few opticians such Poggendorff or Raman on a fundamental point of view but recently applications have emerged in free-space communications, optical trapping, enhanced resolution microscopy, polarimetry... (see [6] for a review) based on transparent and non-chiral crystals. In the case of acentric biaxial crystals, chirality occurs and modifies the conical diffraction. In literature we can find experimental results for α -iodic acid crystal [7] and a few theoretical descriptions [8-9].

Light propagation along or in the vicinity of an optical axis of an acentric and absorbing biaxial crystal leads also to interesting phenomena in the far field. This is so because birefringence vanishes and chirality and linear dichroism are exhibited and even competes. Optical singularities results: C points (circular polarization modes) and L lines (linear polarization modes) [10]. The optical axes are also singular and propagate the Voigt wave [11-13].

This work is devoted to the acentric biaxial Nd doped BZBO laser crystal. We study in the far field the effects of chirality, of linear dichroism and their competition in the 780-940 nm spectral range including the $^4I_{9/2} \rightarrow ^4F_{5/2}$ Nd³⁺ transition from the ground state; in the near field we study the conical diffraction. The experimental results (far field and near field) are interpreted in the framework of a theoretical model of focused polarized-beam propagation including birefringence, dichroism and chirality recently published by one of us [14].

2. Theoretical background

2.1 Propagation of the refractive wave inside an acentric crystal

Inside an acentric absorbing crystal, the propagation of a monochromatic plane wave $\mathbf{E}(\mathbf{r})\exp(i\mathbf{K} \cdot \mathbf{r} - \omega t)$ is governed by a dielectric susceptibility tensor [15-16] written as:

$$\varepsilon^T = \varepsilon_0(\varepsilon + ik_0G) \quad (1)$$

which is the sum of a complex symmetric part ε describing birefringence and linear dichroism (anisotropic absorption) and an hermitic part (G is real antisymmetric, $k_0 = 2\pi/\lambda$) describing the chirality (optical activity).

Eq. (1) is a priori written in the principal dielectric frame xyz , but due to the transverse behaviour of the dielectric displacement vector $\mathbf{D}$, it is convenient to express the tensors in a transverse frame x_1, x_2, x_3 using an underline notation. Then the $\underline{G}$ tensor stands as:

$$\underline{G} = \begin{bmatrix} 0 & -\Gamma & 0 \\ \Gamma & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \quad (2)$$

Γ being the module of the gyration vector calculated from the gyration tensor:

$$\Gamma = g_{ij}u_iu_j \quad (3)$$

where u_i are the coordinates of the unit vector of the propagation direction.

It is a general result (with or without chirality) [11] that the propagation equations can be written as:

$$\frac{d^2}{dx_3^2} \begin{bmatrix} E_1 \\ E_2 \end{bmatrix} = -\frac{\mu_0 \omega^2}{\epsilon_{33}} \begin{bmatrix} \delta_{11} & \delta_{12} \\ \delta_{21} & \delta_{22} \end{bmatrix} \begin{bmatrix} E_1 \\ E_2 \end{bmatrix} \quad (4)$$

introducing a set of δ coefficients. In the case where no chirality occurs the δ coefficients are defined with the symmetric part ϵ of $\underline{\epsilon}^T$:

$$\delta_{11} = \epsilon_{11}\epsilon_{33} - \epsilon_{13}\epsilon_{31} \quad (5a)$$

$$\delta_{12} = \epsilon_{12}\epsilon_{33} - \epsilon_{13}\epsilon_{32} \quad (5b)$$

$$\delta_{21} = \epsilon_{21}\epsilon_{33} - \epsilon_{23}\epsilon_{31} \quad (5c)$$

$$\delta_{22} = \epsilon_{22}\epsilon_{33} - \epsilon_{23}\epsilon_{32} \quad (5d)$$

If chirality is added taking (2) into account inside (1), it is straightforward that the full propagation equations are obtained keeping unchanged the δ_{11} and δ_{22} coefficients and making the change:

$$\delta_{12} \rightarrow \delta_{12} - i\epsilon_0 \epsilon_{33} k_0 \Gamma \quad (6)$$

$$\delta_{21} \rightarrow \delta_{21} + i\epsilon_0 \epsilon_{33} k_0 \Gamma \quad (7)$$

From here in this text the δ coefficients will be strictly defined with the symmetric part ϵ of the susceptibility tensor (that is to say by Eq. 5a-d) and so $\delta_{12} = \delta_{21}$. We can then easily adapt all the formulas established without chirality [17]. First the discriminant of the Fresnel equation corresponding to the general propagation equations Eq. (4) stands as:

$$\Delta_T = \Delta + (2\epsilon_0 \epsilon_{33} k_0 \Gamma)^2 \quad (8)$$

Δ being the discriminant with dichroism alone:

$$\Delta = (\delta_{11} - \delta_{22})^2 + 4\delta_{12}\delta_{21} \quad (9)$$

The x_3 longitudinal component of the complex wave-vector of each of the two modes propagating unchanged along a given direction is obtained as:

$$K_{\pm}^2 = \frac{\mu_0 \omega^2}{2\epsilon_{33}} [(\delta_{11} + \delta_{22}) \pm \Delta_T^{1/2}] \quad (10)$$

It is helpful to introduce the same component $k_{\pm}$ with dichroism alone:

$$K_{\pm}^2 = \frac{k_+^2 + k_-^2}{2} \pm \left[\left(\frac{k_+^2 - k_-^2}{2} \right)^2 + (k_0^3 \Gamma)^2 \right]^{1/2} \quad (11)$$

The transverse components of the eigen-modes are:

$$\begin{bmatrix} 1 \\ \frac{\delta_{22} - \delta_{11} \pm \Delta_T^{1/2}}{2\delta_{12} - 2i\epsilon_0 \epsilon_{33} k_0 \Gamma} \end{bmatrix} \quad (12)$$

The two eigen-modes are elliptically polarized. If the dichroism (symmetric susceptibility tensor) is alone the two ellipse rotations are in the same way, if the chirality is alone (hermitic susceptibility tensor) the rotations are in opposite ways. In the general case of both dichroism and chirality, competition occurs depending of their relative strength.

2.2 C-directions optical singularities

The propagation directions corresponding to the optical axes of the crystal without chirality are of peculiar interest. They are characterized by making $\Delta=0$ in Eq. (8) or $k_+ = k_-$ in Eq. (11) and by $\delta_{22} - \delta_{11} = \pm 2i\delta_{12}$. It is clear that they are no more optical axes in presence of chirality: $\Delta_T \neq 0$ and $K_+ \neq K_-$. However, Eq. (12) leads to the two eigen-modes: $\begin{bmatrix} 1 \\ \pm i \end{bmatrix}$ and

$$\begin{bmatrix} 1 \\ \pm i \frac{\delta_{21} + i\epsilon_0 \underline{\epsilon}_{33} k_0 \Gamma}{\delta_{21} - i\epsilon_0 \underline{\epsilon}_{33} k_0 \Gamma} \end{bmatrix} \quad (\text{the upper sign is valid for propagation in the y-positive half-plane, the lower sign for y-negative}).$$

So, one property of the without-chirality-crystal optical axes remains: one eigen-mode is still circularly polarized, which is an optical singularity, and this direction is so-called a C-direction [10]. The second mode is elliptically polarized.

The C-directions (L and R-C-directions) are easily recognized in the far field pattern working with the crystal sandwiched between two cross circular polarizers: they are extinguished. The second elliptical mode is helpful to test the chirality/dichroism competition: if its ellipse rotation is the same way that the circularly polarized mode, the dichroism dominates. In the opposite situation the chirality dominates. Dichroism and chirality compensates if the ellipse is degenerate into a linear polarization. Then the propagation direction is also a so-called L-direction, so it is twice singular. Inspection of the elliptical eigen-mode shows the compensation occurs if the following relationship is satisfied:

$$|\delta_{21}| = \epsilon_0 \underline{\epsilon}_{33} k_0 \Gamma \quad (13)$$

2.3 Singular optical axes and the Voigt wave

The optical axes are directions with a lone complex wavenumber, i. e. $K_+^2 = K_-^2$. From Eq. (11) it is clear that they disappear if the crystal is transparent (k_+ and k_- are real) and simultaneously chiral ($\Gamma \neq 0$). If the crystal is absorbing we can use complex wavenumbers $k_{\pm} = k'_{\pm} + ik''_{\pm}$ and the conditions for the existence of optical axes in presence of chirality stands as:

$$k_+'^2 - k_-'^2 = k_+''^2 - k_-''^2 \quad (14)$$

$$k_+ k_+'' - k_- k_-'' = \pm k_0^3 \Gamma \quad (15)$$

Eq. (15) means that if the linear dichroism represented by the (15) left side is not high enough whatever the propagation direction, no optical axis exists and the chirality dominates. If linear dichroism is high enough Eq. (14) and (15) can be fulfilled in some directions and optical axes exist.

In such a case the optical axes are singular and propagate the Voigt wave:

$$\begin{bmatrix} E_1(L) \\ E_2(L) \end{bmatrix} = \left\{ \begin{bmatrix} E_1(0) \\ E_2(0) \end{bmatrix} + iKL \frac{\delta_{12} - i\epsilon_0 \underline{\epsilon}_{33} k_0 \Gamma}{\delta_{11} + \delta_{22}} \begin{bmatrix} 1 \\ \pm i \left(\frac{\delta_{12} + i\epsilon_0 \underline{\epsilon}_{33} k_0 \Gamma}{\delta_{12} - i\epsilon_0 \underline{\epsilon}_{33} k_0 \Gamma} \right)^{1/2} \end{bmatrix} \begin{bmatrix} E_1(0) \\ E_2(0) \end{bmatrix} \right\} e^{iKL} \quad (16)$$

We can see that due to chirality the Voigt wave polarization (second term of the Eq. (16) right side) is elliptical.

2.4 Far field pattern

The electric field $\mathbf{E}$ of the input beam at the entrance face of the crystal is a priori known in the (x', y', z') frame represented in the onset in Fig. 1, z' being the direction of the optical axis of the transparent crystal (removing chirality) and orthogonal to the entrance crystal face. This field can be decomposed in plane waves from its 2D-Fourier transform $\hat{\mathbf{E}}$. Each plane wave propagates inside the crystal after refraction according to a calculation detailed in Ref. [14] and summarized as:

-step 1: we obtain the $\hat{\mathbf{E}}$ components and the dielectric susceptibility tensor components $\underline{\varepsilon}^T$ in the (x_1, x_2, x_3) transverse frame

-step 2: with the help of the eigen-modes we obtain the $\hat{\mathbf{E}}$ tangential components at the output crystal face

-step 3: we go back to the (x', y', z') frame which leads to the electric far field pattern.

2.5 Near field pattern

After propagation in air over a distance d behind the crystal, all the plane waves constituting the far field interfere, in other words the near electric field is obtained as an inverse 2D-Fourier transform iFT . From discretisation the inverse Fourier transform is calculated with the inverse Fast Fourier Transform (iFFT) algorithm of the Matlab package.

2.6 The Nd-doped BZBO crystal

The BZBO crystal is orthorhombic with point group $mm2$ (C_{2v}). The x, y, z axes of the dielectric frame coincide respectively with the b, c, a crystallographic axes. So the optical axis of the transparent crystal lies in the xz -ba plane with the V_z angle with the z axis close to 42.6° , depending slightly on the wavelength. In the xyz frame the complex ε tensor of the Nd-doped BZBO is diagonal:

$$\varepsilon = \begin{bmatrix} n_x^2 + i \frac{n_x \lambda}{2\pi} k_x'' & 0 & 0 \\ 0 & n_y^2 + i \frac{n_y \lambda}{2\pi} k_y'' & 0 \\ 0 & 0 & n_z^2 + i \frac{n_z \lambda}{2\pi} k_z'' \end{bmatrix} \quad (17)$$

where k_x'', k_y'', k_z'' are the absorption coefficients which are twice the imaginary part of the complex wavenumbers.

The two mirrors of the crystallographic structure are the yz and the xy planes, so the lone non-vanishing components of the gyration tensor are $g_{xz}=g_{zx}$. The module of the gyration vector is:

$$\Gamma = 2g_{xz}u_xu_z \quad (18)$$

and is non-vanishing along propagation directions near the optical axis; optical activity is then expected.

3. Experimental methods

BZBO melts congruently near 690°C . There are no phase transitions between the temperature of crystallization and the room temperature, therefore crystal growth from melt of

stoichiometric composition can be carried out. The Czochralski method looks to be the method of choice to obtain bulk single crystals of BZBO for optical investigations. However, the relatively high viscosity of molten BZBO makes it very difficult to ensure stable crystal-melt interface when pulling up of the growing crystal is used. Addition of RE ions further worsens growth conditions, segregation of dopants on the interface leads to constitutional supercooling and formation of inclusions. Taking the above into account we decided to carry out BZBO:Nd crystallization by means of Kyropoulos method under conditions of low temperature gradients, similarly to crystallization of NLO $\pm$ -BiB₃O₆ [17]. BZBO single crystals grew on oriented seeds in the volume of the melt. No pulling was used, growing crystals were confined with crystallographic planes. Crystallization was carried out from a platinum crucible 40 mm in diameter and 40 mm in height. Two-zone resistance furnace controlled with Eurotherm 906S programmers ensured stable thermal conditions and axial temperature gradient of about 1 K/cm. The temperature of the melt was lowered at a rate of 0.01K/h during crystallization, growing crystal was rotated at a rate of 20 rpm. After the withdrawal of the as-grown BZBO crystal from the melt it was cooled down to the room temperature at a rate of 6 K/h.

The xyz-polarized absorption spectra were recorded from two y-cut and z-cut Nd-doped BZBO samples with a Perkin Elmer 900 spectro-photometer.

A third optical axis-cut sample with 0.538 cm thickness was used for focused beam propagation and imaging. It was inserted in a holder equipped for rotation around three orthogonal axes (one longitudinal and two transverse).

The CW beam used for far and near fields imaging was provided by a Coherent Titane-sapphire laser pumped by a frequency doubled Nd:YAG laser at 532 nm. Its wavelength was tunable from 770 up to 940 nm with 0.01 nm spectral width. The beam, focused slightly behind the sample (1 cm) with a 5 cm focal length lens L1, is schematically represented in Fig. 1. Then it is separated in two parts by a 50/50 beam splitter towards two CCD cameras from Ocean Optics. The near field (red circle resulting of conical diffraction behind the crystal) is imaged through the L2 lens (15 cm focal length) with a magnification 6.9 on the CCD1 screen, the magnification being calculated from the lens-camera distance. The far field is obtained through the L3 lens (25 cm focal length) on the CCD2 screen located at 25 cm from the L3 lens. All the lenses are achromatic doublets from Thorlabs.

In Fig. 1 we can see that a linear polarizer and an achromatic quarter-wave plate (from Thorlabs) can be introduced in the beam path before the sample in order to make the beam polarized linearly, circularly or elliptically. After the sample similar optical components are used to analyse the beam and decompose it in different polarization states.

Fig. 1 Experimental set-up for beam propagation through the Nd:BZBO optical-axis oriented sample and for far and near fields imaging.

4. Experimental results and interpretation

4.1 Crystal growth

The habit of as-grown BZBO single crystals strongly depended on the orientation of the seed. It was found that [001] seed orientation was most appropriate to grow uniform bulk crystals, while [100] or [010] seed directions resulted in growth of flat BZBO crystals elongated in [001] direction. The morphology of as-grown [001] BZBO:Nd single crystal is described below. Fig. 2 shows as-grown BZBO:Nd single crystal. Lateral crystallographic planes confining the crystal can be seen. Dark spots were formed on the surface of the crystal by droplets of melt that turned black-brown when solidified.

Fig. 2. As-grown [001] BZBO:Nd single crystal.

Fig. 3 shows morphology of the BZBO:Nd single crystal in direction perpendicular to (001) plane. First $\{100\}$, $\{010\}$, and $\{111\}$ planes were identified using method described in [18]. Taking into account measured and calculated angles between particular planes and directions, as well as results of crystallographic zone calculus, full morphology of the as-grown BZBO:Nd single crystal was obtained.

Fig.3 View of formed crystallographic planes of [001] BZBO single crystal described by Miller indices for (hkl) planes.

Orientation of the samples for optical investigations was made by means of the Laue method and analytical use of the stereographic Wulff net [19].

4.2 Absorption

The absorption spectra in x, y, z linear polarizations corresponding to the $^4I_{9/2} \rightarrow ^4F_{5/2}$ Nd^{3+} transition from the ground state are represented in Fig. 4 a. The absorption is strongly anisotropic, it is much higher for polarization in the xz principal plane than perpendicularly. The absorption coefficients lead to the knowledge of the ϵ symmetric tensor (Eq. (17) adding the wavelength dependence of the refractive indices measured in Ref. [3].

The linear dichroism understood as the difference between the absorption of the two modes propagating unchanged in a given direction has its maximum value in the xz-principal plane and we have calculated it for the direction of the optical axis of the transparent crystal without chirality. This direction makes the angle V_z (about 46.2° depending slightly on wavelength) with the z-axis. The linear dichroism is obtained as $(k_x'' \cos(V_z)^2 + k_z'' \sin(V_z)^2) - k_y''$. It is represented versus wavelength in Fig. 4 b.

Fig. 4 (a) Polarized absorption spectra for the $^4I_{9/2} \rightarrow ^4F_{5/2}$ transition in Nd:BZBO; (b) Linear dichroism for the optical axis (see text) propagation direction.

4.3 Chirality and conical diffraction in the spectral domain of transparency

In this sub-section we restrict the study at wavelengths of crystal transparency slightly outside the $^4I_{9/2} \rightarrow ^4F_{5/2}$ Nd³⁺ transition. For example in Fig. 5 all the images have been recorded at 780 nm. The experimental far field pattern obtained inputting a right circular polarized focused beam in the crystal and selecting at the output a left circular polarization (R-L configuration) is represented in Fig 5 (a1). The central extinguished point is interpreted as a C-point (section 2.2) and not as an optical axis due to chirality. The latter affirmation is proved inputting a vertical polarized light and selecting an output horizontal polarization (Fig. 5 (b1)). The central point is no more extinguished (as it is the case with a non-chiral crystal). In fact the central direction can be fully extinguished rotating the linear output polarizer, meaning that the light propagation along the central direction remains linearly polarized with a rotation of its polarization measured to be 50°, that is to say 92.9°/cm specific rotatory power (rotation angle per unit path inside the crystal). At 850, 860, 870, 890, 920, 940 nm the rotation angles were measured respectively as 43°, 40°, 33°, 32°, 30° and 27°, corresponding to 79.9, 74.3, 61.3, 59.4, 55.5 and 50.2°/cm specific rotatory power.

An elementary calculation involving two left and right circular polarizations constituting a linear polarized wave shows that in this C-point direction the Γ_{C-P} module can be obtained from the specific rotatory power ρ by:

$$\Gamma_{C-P} = -\frac{\lambda^2 n_2}{2\pi^2} \rho \quad (18)$$

and finally from Eq. (3):

$$g_{xz} = \frac{\Gamma_{C-P}}{2 \sin(V_z) \cos(V_z)} \quad (19).$$

At this step the dielectric susceptibility tensor (1) is fully determined (with extrapolation between measured wavelengths) and the far and near fields patterns can be calculated from section 2.

The theoretical patterns corresponding to the experimental (a1) and (b1) are represented in Fig. 5 by (a3) and (b3) respectively; they are in reasonable agreement.

Fig. 5 Lines 1 and 3: experimental and theoretical far field; Lines 2 and 4: experimental and theoretical near field; Column (a): R-L configuration; Column (b): V-H configuration; Column (c): R-V configuration; Column (d): R-45° configuration.

Calculation of the near field pattern needs the knowledge of the input Gaussian beam diameter. We have used the measured value: 24 μm .

The near field patterns, measured and calculated in the plane of focalisation, visualize rings due to conical diffraction in Fig. 5 (a2) and (a4) for R-L configuration and (b2) and (b4) for V-H configuration. In the latter case, the ring is not symmetric: the lower part is more intense, this is due to the chirality induced polarization rotation.

We have selected other polarizer/analyser configurations to point out the effects of chirality. Spirals already encountered in α -iodic acid crystal [7-8] exist also for the BZBO crystal symmetry. In Fig. 5 the four pictures of column (c) correspond to the D-V configuration and the picture of column (d) to the D-45° configuration. Let us notice the slight fuzziness of two horizontal ((c1) and (c3)) and vertical ((d1) and (d3)) strips superimposed on the spirals.

4.4 Chirality versus linear dichroism competition in the absorption domain

The chirality/dichroism competition was first studied from propagation along the C-directions by inspection of the $\begin{bmatrix} 1 \\ tg(\chi)e^{i\phi} \end{bmatrix}$ elliptical mode (expressed from sub-section 2.2) going with the circular one. The laser wavelength was fixed successively at the peak or valley values of the absorption spectra in Fig. 4 (a) ordered by growing absorption: 820.4, 794.3, 813.0, 804.8, 801.6 and 808.0 nm. For each wavelength the L- C-directions in the xz upper half-plane and the R-C-directions in the xz lower half-plane are easily found in the far field (using two opposite circular polarizer and analyser) as the extinguished point slightly above or below the centre of the pattern (see Fig. 6 (a1) for example). The characteristics of the elliptical modes (ellipticity ϵ and α larger axis angle with horizontal axis) were calculated and gathered in Table I. This mode was generated experimentally with an elliptical polarizer (playing with a linear polarizer and a quarter wave-plate) before the crystal. A crossed elliptical analyser behind is used to check light extinction. The latter was obtained, only a few cases required a slight adjustment of the ellipticity. We can see in Table I that for a given C-direction, at any wavelength except 808.0 nm, the circular and elliptical eigen-modes are always in opposite rotation (opposite ϕ signs), meaning that chirality dominates. This is the contrary at 808.0 nm absorption peak (same ϕ signs), the dichroism dominates.

We have examined the exact balance between chirality and dichroism by studying Eq. (13) versus wavelength in the absorption spectra. We have found that the elliptical mode degenerates in the linear $\begin{bmatrix} 1 \\ -1 \end{bmatrix} / \sqrt{2}$ mode at 806.6 and 809.2 nm, these two wavelengths being marked in Fig. 4 (a).

The other aspect of the chirality/dichroism competition is the existence of the optical axis studied in sub-section 2.3. Of course the conditions (14) and (15) can be exploited through their exact solutions but here we will content ourselves with the approximation that the imaginary parts of the wave-numbers are weak by comparison of the real parts, so $k'_+ \cong k'_-$ from (14). In this case the left side in Eq. (15) is the linear dichroism which has to be calculated in the propagation directions leading to its highest value. We will use the result anticipated in sub-section 4.1 where it was calculated for the direction of the optical axis of the transparent crystal without chirality. The right side of Eq. (15) is then $k_0^2 \Gamma / n_2 = 5.8 \text{ cm}^{-1}$. This value is reported in Fig. 4 (b) as a horizontal red line visualizing the border separating the two chirality or dichroism domination spectral regions corresponding to absence or existence of the singular optical axes. Within our approximation the borders in Fig. 4 (a) and (b) are very close.

Fig. 6 Lines 1 and 2: experimental and theoretical fields at 809.6 nm; Column (a): R-L configuration far field; Column (b): Extinction of the linear eigen-mode of the optical axis in the far field; Column (c): Voigt wave transmission in the far field; Column (d): same configuration than column (c) in the near field;

Finally we have fixed the wavelength to the 809.2 nm value realizing the balance chirality/dichroism. The C-direction obtained in the far field in L-R configuration is shown extinguished slightly above the centre of Fig. 6 (a1) (experimental) and 6 (a2) (theoretical). The optical axis is located on the xz principal plane, so $\delta_{12} = 0$ in its direction and the Voigt wave polarization (Eq. (16)) is linear: $\begin{bmatrix} 1 \\ -1 \end{bmatrix} / \sqrt{2}$. This is experimentally checked by inputting this linear polarization in the crystal. The first term in the right hand side of (16) is non-zero and transmitted unchanged, the second term is zero. Using a cross linear polarizer behind the crystal, the centre of the far field pattern is extinguished (Fig. 6 (b1) (experimental) and 6 (b2) (theoretical)). To the contrary, if we input a linear polarization orthogonal to the Voigt wave (i. e. $\begin{bmatrix} -1 \\ 1 \end{bmatrix} / \sqrt{2}$), a cross linear polarizer behind the crystal will transmit the Voigt wave (and only this wave) and the centre of the far field pattern appears brilliant (Fig. 6 (c1) (experimental) and 6 (c2) (theoretical)). The corresponding near field pattern in the focal plane of the transmitted Voigt wave is represented in Fig. 6 (d1) (experimental) and 6 (d2) (theoretical).

5. Conclusion

We have grown acentric chiral Nd-doped BZBO single crystals by the Kyropoulos method under conditions of low temperature gradients. Samples were oriented and cut perpendicular to the optical axis located in the ab plane at 42.6° from the a-axis. We have focused slightly behind the crystal a Sa-Ti laser beam at selected wavelengths in the spectral domain of transparency (780, 850, 860, 870, 890, 920, 940 nm) with various linear or circular polarizations. The output far field was projected on various polarization states in order to visualized conoscopy patterns revealing the specific role of chirality: disappearance of the optical axis replaced by two C-directions, linear polarization rotation and spiral patterns. One main result is the measure of the specific rotatory power. On the other hand, the output near field exhibits the influence of chirality on the conical diffraction. Going to selected wavelengths in the spectral domain of absorption corresponding to the $^4I_{9/2} \rightarrow ^4F_{5/2}$ Nd³⁺ transition, we

first studied the chirality/dichroism competition from propagation along the C-directions, characterizing the circular and elliptic eigen-modes. We exhibited the wavelengths at which the ellipse degenerates in a linear polarization, signature of the chirality/dichroism balancing. The second aspect of the chirality/dichroism competition is the existence of the optical axis which can be restored if the dichroism is high enough. In this case a singular Voigt wave can propagate. From the experimental determination through absorption and optical activity of the optical parameters, a theoretical description has been proposed, leading to quite well agreement in the far field (conoscopy) and in the near field (conical diffraction).

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20. λ (nm)	χ (°)	ϕ (°)	α (°)	ε (°)	Domination
820.4	45	90 (L)	-	45	Chirality
	45	-102.2 (R)	-45	38.9	
	45	-90 (R)	-	45	
	45	102.2 (L)	-45	38.9	
794.3	45	90 (L)	-	45	
	45	-109.2 (R)	-45	35.4	
	45	-90 (R)	-	45	
	45	109.2 (L)	-45	35.4	
813.0	45	90 (L)	-	45	
	45	-126.8 (R)	-45	26.6	
	45	-90 (R)	-	45	
	45	126.8 (L)	-45	26.6	
804.8	45	90 (L)	-	45	
	45	-147.7 (R)	-45	16.1	
	45	-90 (R)	-	45	
	45	147.7 (L)	-45	16.1	
801.3	45	90 (L)	-	45	
	45	-162.0 (R)	-45	9	
	45	-90 (R)	-	45	
	45	162.0 (L)	-45	9	
808.0	45	90 (L)	-	45	Dichroism
	44.95	148.8 (L)	-44.94	15.5	
	45	-90 (R)	-	45	
	45.04	-148.8 (R)	-45.05	15.5	

Table I Characterization of the circular and elliptic eigen-modes in the C-directions determined by the selected wavelengths in the absorption domain.

Figure captions

Fig. 1 Experimental set-up for beam propagation through the Nd:BZBO optical-axis oriented sample and for far and near fields imaging.

Fig. 2. As-grown [001] BZBO:Nd single crystal.

Fig.3 View of formed crystallographic planes of [001] BZBO single crystal described by Miller indices for (hkl) planes.

Fig. 4 (a) Polarized absorption spectra for the $^4I_{9/2} \rightarrow ^4F_{5/2}$ transition in Nd:BZBO; (b) Linear dichroism for the optical axis (see text) propagation direction.

Fig. 5 Lines 1 and 3: experimental and theoretical far field; Lines 2 and 4: experimental and theoretical near field; Column (a): R-L configuration; Column (b): V-H configuration; Column (c): R-V configuration; Column (d): R-45° configuration.

Fig. 6 Lines 1 and 2: experimental and theoretical fields at 809.6 nm; Column (a): R-L configuration far field; Column (b): Extinction of the linear eigen-mode of the optical axis in the far field; Column (c): Voigt wave transmission in the far field; Column (d): same configuration than column (c) in the near field.