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Wavelength dependence of the orientation of optic axes in KGW

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Philippe Veber · Todor K. Kalkandjiev · Daniel Rytz ·
Lionel Canioni · Marc Eichhorn

Abstract We report on the wavelength dependence of the orientation of the optic axes in monoclinic crystals using undoped and neodymium-doped $\text{KGd}(\text{WO}_4)_2$ (KGW) samples. Measurements have been performed over a wide wavelength range from 420 up to 1,580 nm with more than 70 measurement points. This provides the widest measurement range of the optic axis variation ever reported in literature for KGW crystals. With such a measurement, which can be performed in principle in all biaxial crystals, the refractive indices values can be checked with a precision down to 10^{-3} .

1 Introduction

An old effect commonly called internal conical refraction (ICR) is attracting more and more interest in the last few

years. It has been first predicted by Hamilton in 1832 and quickly observed by Lloyd [1] few months later. Several additional contributions improved the knowledge of this effect, with the observation of the secondary ring [2] and axial spike [3]. Since its prediction by Hamilton, several papers have improved and refined the theory of ICR [4–10]. Furthermore, the authors [11] have analyzed the role of ICR on second harmonic generation in biaxial crystals. The theoretical aspect has been recently revisited by Berry in several papers [12, 13]. From the theory, one can predict the beam evolution in biaxial crystals. Even the theoretical description of cascaded ICR is reported [14]. ICR appears when a light beam propagates along one of the optic axes. Considering potential applications of this phenomenon, the formation of the characteristic ring by ICR gives access to beam shaping [15–18] and offers new possibilities for innovative configurations in molecule trapping or for some micromachining applications. Even the use of this effect in laser gain media has been explored [19–21]. From those last articles, it results that the orientation of the crystal is important and should be controlled with an accuracy of a few mrad. At present, the most studied material for ICR is $\text{KGd}(\text{WO}_4)_2$ (KGW). This choice can be explained by the available good crystal quality of this host matrix and its strong anisotropy compared with other biaxial crystals that are accessible with sufficient quality. The variation of the optic axis orientation in space for different wavelengths can be easily calculated from the refractive indices. However, the precision of the three refractive indices must be very accurate to predict those angles. Unfortunately, it is not easy to determine the three principal refractive indices of biaxial crystals. Several studies have been performed on different biaxial crystals in order to characterize the wavelength dependence of their optical properties. However, the low number of measurement points reduces the

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precision and thus limits the prediction of the results [11, 22–24]. One publication provides impressive results on the wavelength dependence of refractive indices for the monoclinic crystal BIBO [25]. Their measurements of refractive indices were used to calculate the variation of the optic axes with accuracy. However, to the best of our knowledge, no experiments have been performed so far in order to confirm this evolution. In the present paper, we experimentally investigate the wavelength dependence of the optic axes for Nd-doped and undoped KGW within a large wavelength range from 420 to 1,580 nm, and we compare our results with the ones calculated from refractive indices reported in literature [26]. We suggest a small modification of the values of the indices reported in the literature to obtain an satisfactory agreement between calculated and experimentally determined optic axis orientations.

2 Optic axes

In biaxial crystals, there are two optic wave axes, located in the XZ plane of the indices surface. The directions X , Y and Z represent the axes of the indicatrix having three principal refractive indices n_x , n_y and n_z , respectively, where $n_x < n_y < n_z$.

This optical coordinate system is orthogonal and does not coincide with the crystallographic axes. However, in monoclinic KGW crystals, the direction X corresponds to the crystallographic axis $\mathbf{b}$ and the ZY and $\mathbf{a}\mathbf{c}$ planes are the same. To set the direction of this optical coordinate system, the angle ϕ , as shown in Fig. 1, must be known and may depend on temperature and wavelength.

Using X-ray diffraction, Laue patterns of the crystals have been recorded with a CCD camera from Photonic Science for a crystal orientation perpendicular to the entrance facet. The samples were aligned parallel on one plate together with a crystal of silicon. By using the silicon Laue pattern as calibration reference, the plate was perpendicularly aligned to the X-ray beam coming from the

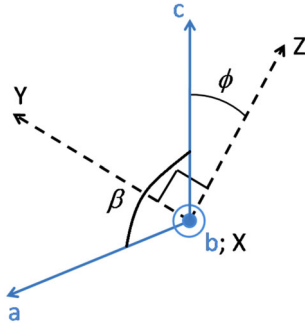


Fig. 1 Orientation of the crystallo optical coordinate with respect to crystallographic axes

molybdenum anti-cathode. Using analyzing software based on the Laue method (Orient Express), we could determine all angles between crystallographic axes with respect to our crystal orientation. The mechanical alignment of our samples, relative to the silicon crystal, is the main source of uncertainty. For this purpose, the unit cell parameters of Pujol et al. [26], which are $\mathbf{a} = 10.652 \text{ \AA}$, $\mathbf{b} = 10.374 \text{ \AA}$, $\mathbf{c} = 7.582 \text{ \AA}$ and $\beta = 130.80^\circ$, were used. Knowing that for those monoclinic crystals the principal axis of n_p (X) corresponds to the crystallographic axis $\mathbf{b}$, we found that the angle ϕ between the direction of n_z (Z) and the crystallographic axis $\mathbf{c}$ was about $20.0^\circ \pm 1$ and $20.7^\circ \pm 1$ for undoped KGW and Nd-doped KGW, respectively. Furthermore with those, records references of angles V at 633 nm were found to be 43.3° and 42.1° , respectively. Note that in case of the undoped crystal, a correction has been added in order to take into account the misalignment of this sample at 633 nm (see next section). The main uncertainty of this method (which is also the main source of uncertainties in our measurements) results in the positioning of the crystal on the translation stage which needs to be perpendicular to X-rays. All those angles calculation is simplified due to monoclinic symmetry. In case of triclinic crystals that are also biaxial, it will be more difficult to determine them.

The angle V , between the n_z (Z) axis and the optic axis in the XZ plane, can be easily calculated:

$$\tan^2(V) = \frac{n_z^2(n_y^2 - n_x^2)}{n_x^2(n_z^2 - n_y^2)} \quad (1)$$

Equation 1 is given without any approximation.

Note that in this paper, we discuss only about the optic wave axes (sometimes called binormal) from the index surface (called wave surface in [27]), along which one the ICR takes place. It exists another optic ray axes (also called biradial) from the ray surface. Similar angle V_r can be calculated replacing n_i by $\frac{1}{n_i}$ in Eq. 1 with $i = x, y, z$.

In this work, the theoretical angle between the principal axis corresponding to n_z and the optic axis given by Eq. 1 is used to compare the calculated wavelength dependence of V with the experimental results of the optic axis direction shift at room temperature. The three principal refractive indices of KGW can be found in the literature in papers by Pujol et al. [26] for Er-doped KGW and by Konstantinova et al. [28] for Nd-doped KGW. Their values are depicted in Fig. 2. The refractive indices from Pujol et al. [26] are given with an accuracy of 10^{-3} from 400 to 1,600 nm and those from Konstantinova et al. [28] with an accuracy of 5×10^{-4} from 430 to 640 nm. As one can see on Fig. 2, the values of n_x and n_y are similar for both authors in their measurement ranges. Only the values for n_z do not coincide for the two crystals, with a lower value observed for Nd-doped KGW.

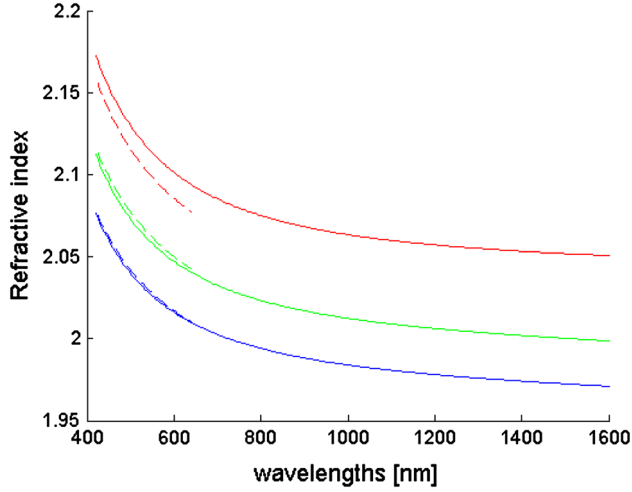


Fig. 2 Refractive index evolution of the three principal indices of Er doped (*solid line*) and Nd doped (*dashed line*) KGW taken from [26, 28]

3 Experimental setup

The measurements of the angular variation of the optic axis orientation with wavelength were performed at LOMA in Bordeaux. The range from 430 up to 1,580 nm could be covered using a Ti:Sapphire laser, a compact OPO (Chameleon/Coherent) and optional frequency doubling with a BBO crystal. The wavelength of the primary Ti:Sapphire source is tunable from 680 to 1,080 nm. With the OPO, we were able to reach the wavelength range from 1,040 up to 1,580 nm. By doubling the Ti:Sa laser, the wavelengths from 430 to 530 nm, and by frequency doubling of the output beam after the OPO, 520–670 nm were obtained. To ensure very precise alignment of the incident beam on the crystals for all wavelengths, the beam of the selected wavelength was injected into a single-mode optic fiber. Two different fibers have been used for the visible and the infrared range, Thorlabs P1-460A-FC-2 and P1-630A-FC-2, respectively. A collimated output FC connector was used to ensure that the output beam will point exactly in the same direction even when the fibers are switched. A lens ($f = 125$ mm) was used to focus the light inside the crystals, another one ($f = 125$ mm) was used to more or less collimate the beam after the crystal, and a last one of short focal length ($f = 40$ mm) was used to magnify the rings for better observation, and to correct the focal variation with wavelength. Figure 3 shows a schematic drawing of the experimental setup. The two Nd-doped (at 3 at.%) KGW samples with dimensions $3 \times 4 \times 11$ mm and an undoped KGW crystal with the dimensions $3 \times 3 \times 10$ mm were used. Each of the samples was fixed on a crystal mount which was positioned on a goniometer driven by a microcontroller with an angular step of 10^{-3} degrees.

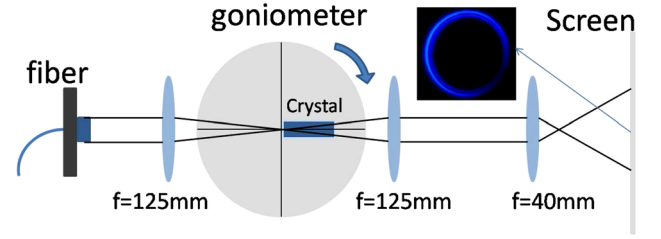


Fig. 3 Scheme of the experimental setup

The sample mount was very accurately fixed in the center of the goniometer. The doped crystals were cut for ICR at 633 nm with a misalignment of 1.25 mrad. Therefore, the first alignment for ICR was performed at this wavelength for one of the doped crystals by turning the goniometer until the CR rings appeared.

To ensure that the plane of variation of the optic axis was perpendicular to the rotation axis of the goniometer, we applied a simple and efficient alignment procedure. The wavelength was changed, and the goniometer was turned until the ICR was aligned in the vertical rotation axis. Instead of tilting the crystal with the horizontal rotation axis to have correct alignment, the crystal was rotated with respect to the propagation axis. To check the alignment, an upper and a lower wavelength several hundred nanometers apart were set successively and we verified that we only turn the goniometer to re-obtain the rings for both wavelengths. With this method, we ensure that the horizontal plane of the crystal is well aligned in its XZ plane.

The second Nd-doped crystal was then aligned with back reflection, whereas the undoped KGW was the most difficult to align in the right plane due to its large cutting imprecision which leads to a deviation of 70 mrad. This difference can be explained by the fact that the crystals were bought from different companies.

As not all wavelengths were available simultaneously because of the switching from the Ti:Sapphire laser to the OPO, realignment of the fiber injection was necessary. We ensured correct reproducibility by checking the overlap for some wavelengths accessible with the different sources.

4 Results

The angular variation obtained with this measurement system for KGW and Nd-doped KGW is shown in Fig. 4 with a comparison to the calculated optic axis angle from the refractive indices of Er-doped KGW and Nd-doped KGW using Eq. 1. It is important to note that the measurements performed with this method only give the relative variation of the angle. In order to compare the result with calculated values, the reference angle of 43.3° and 42.1° at 630 nm has been chosen to scale the experimental

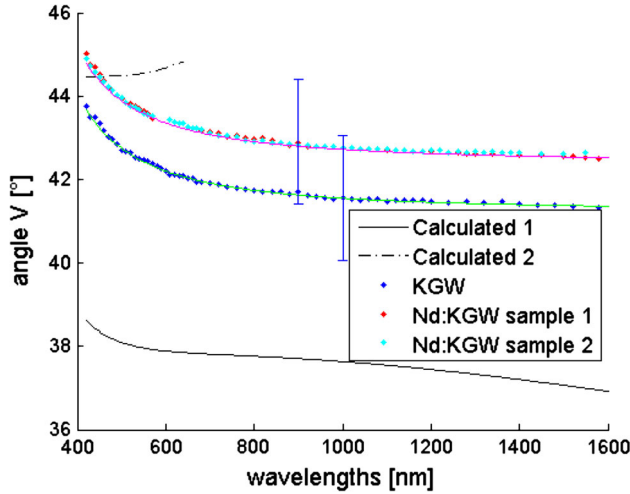


Fig. 4 Angle V as a function of the wavelength (calculated values 1 and 2: obtained using Sellmeier equations from Pujol et al. [26] and from Konstantinova et al. [28], respectively, measurement of the angle V variation is referenced at 43.3° and 42.1° at 630 nm for Nd doped KGW and undoped KGW, respectively; green and pink curves are calculated using the modified n_z refractive index)

values of Nd-doped KGW and undoped KGW, respectively. Those reference angles have been determined using X-ray measurements of the crystals. The measured variation angles of the goniometer scale have been divided by n_y , to take into account the refraction at the entrance surface.

We observe a variation of the optic axis that becomes asymptotic in the infrared region. The variation within the range of 430–1,580 nm is about 2.4° . No significant difference of the curve shape is observed between the doped and undoped double tungstates. Some measurement points could not be taken for the doped KGW crystals, because of the absorption in the visible region of these samples. For example at 600 nm, we should consider if those absorption bands could lead to a change of the birefringence and thus influence the dispersion. However, the highest variation of the birefringence due to absorption reported [28] is about 10^{-6} and located at 600 nm. This is far too low to have a significant impact on the optic axis orientation and can thus be neglected.

Moreover, over the entire investigated spectral domain, we did not observe any rotation of the index ellipsoid along the crystallographic b -axis [corresponding to the n_x (X) direction] like it was demonstrated in other monoclinic hosts [25]. During the measurement process, only a single screw was used to align successively each wavelength for ICR. The orientation changes observed in KGW therefore can only occur in one plane within our wavelength measurement range and within the mechanical accuracy. A rotation in the other plane may exist on a very small level or for an extended measurement range.

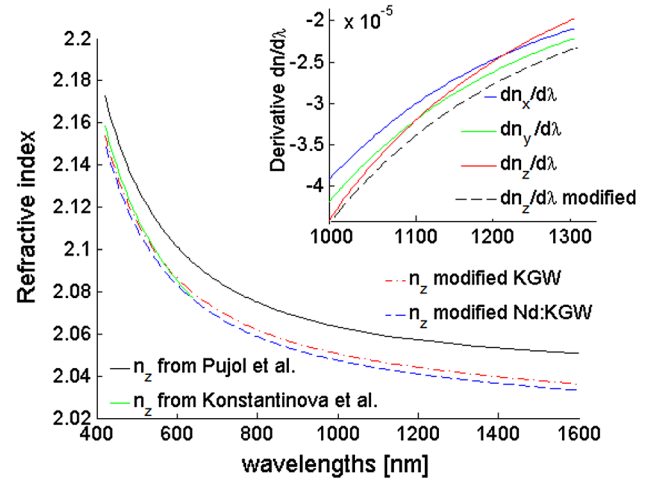


Fig. 5 Derivative of Pujol et al.'s refractive index and the modified one (inset, top right) and comparison between n_z from Pujol et al. and the modified ones (see text)

The measurements were reproducible with an uncertainty of around 0.1° in relative measurements. The uncertainty of the reference used is much higher and was determined by the X-ray diffraction measurement to approximately 1.5° (indicated by the error bars in Fig. 4). For some points, the relative uncertainty is slightly higher, due to the difficulty to align for ICR with low intensity. This problem occurs beyond 1,500 nm, and for some absorption regions of the Nd-doped samples in the visible range. However, one can observe that the shapes of the measured and the theoretical curves are significantly different in the infrared region. This arises from the extreme sensitivity of the angle V on the refractive index value differences. In fact, a slight change of only one of the three refractive indices values changes the shape of the calculated graph and allows for a good agreement in the two curves. In the next paragraph, such a correction of the refraction index is shown. As can be seen in Fig. 4, the prediction calculated from the indices of reference [28] does not match our experimental result at all. This is probably due to Sellmeier fitting performed over a limited wavelength range with a limited number of data points only.

5 Modification of the refractive index " n_z "

In this part, a correction will be applied to the refractive index of Pujol et al. [26] in order to bring the calculated values into agreement with the measured ones. This kind of correction cannot be applied on Konstantinova et al.'s refractive indices [28] due to the limited measurement range which does not allow us to determine an evolution of one refractive index. Using Eq. 1, it is possible to define one refractive index as a function of the angle V and of the two other refractive indices. This will be used to

Table 1 Sellmeier's coefficients of KGW's n_z refractive index, with C in nm and D in $\times 10^{-9} \text{nm}^{-2}$

Coefficients	A	B	C	D
Pujol et al.	1.3867	0.6573	170.02	0.2913
Modified KGW	1.535	0.5002	184.4	2.233
Modified KGW:Nd	1.535	0.4971	184.8	2.194

recalculate one of the 3 refractive indices with the means of the measured angle V . The n_z refractive index is the one that will be corrected for two reasons. The first one is that as shown in Fig. 2, n_x and n_y seem to be equal for differently doped KGW matrices. The second reason is illustrated in Fig. 5. In fact, looking at the derivatives $\frac{dn}{d\lambda}$ of the indices of Pujol show that n_z does not follow the same variation as n_x and n_y . It is this difference that leads to the variation of the calculated angle V in Fig. 4. This perturbed derivative evolution could be linked to the last measurement point of n_z near 1,500 nm in Pujol et al. [26] which is not well matched by the Sellmeier fit.

In Fig. 5, the modified n_z values with respect to the original ones from Pujol et al. are shown for comparison. The difference between Pujol et al. n_z refractive indices and the refined ones is about 0.014 and 0.018 for undoped KGW and Nd-doped KGW, respectively. Therefore, a small correction of the refractive index n_z can be derived from our experimental results. The index is given by Eq. 2.

$$n_z = A + \frac{B}{1 - \left(\frac{C}{\lambda}\right)^2} - D\lambda^2, \quad (2)$$

where the Sellmeier coefficients corresponding to Eq. 2 are given in Table 1.

With this new refractive index, the angle V can be recalculated. Indeed, the angle V calculation is very sensitive to the refractive index, for a variation of the refractive index of 0.0005, the angle may vary by 0.15° . With our Sellmeier fit, it is not possible to be more precise than this value.

6 Conclusion

We have shown that in order to predict the evolution of the optic axis with wavelength, one must know the three refractive indices values to better than 10^{-3} . The variation of the angle V has been measured over the range from 430 to 1,580 nm, and a correction of the n_z refractive index taken from literature has been performed. Over the measurement range, a variation of 2.4° has been measured. No significant difference of the variation has been observed

between doped and undoped KGW materials. This kind of measurement can be used to check the precision of the refractive index at the 10^{-4} level with respect to an angular accuracy of the optic axis of 0.01° . Furthermore, no rotation of the index ellipsoid around the crystallographic b-axis has been observed.

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