

Refined orientation of the optical axes as a function of wavelength in three monoclinic double tungstate crystals $\text{KRE}(\text{WO}_4)_2$ (RE = Gd, Y or Lu)

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Abstract: We measured a rotation of the dielectric frame as a function of wavelength of about 2° in the spectral range of 0.4–1.6 μm in the monoclinic double tungstate crystals, $\text{KRE}(\text{WO}_4)_2$ where RE = Gd, Y or Lu. We also refined the Sellmeier equations of their three principal refractive indices. By taking into account these data, we calculated the real orientation of the optical axes as a function of wavelength which is of key importance for the design of conical refraction lasers based on monoclinic double tungstate crystals.

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1. Introduction

Double tungstate (DT) crystals with the chemical formula $KRE(WO_4)_2$ (where RE denotes a "passive" ion like Gd, Y or Lu) labelled KGdW, KYW and KLuW, are well-known. When doped with trivalent lanthanide ions, *e.g.* Yb^{3+} or Tm^{3+} , they provide intense and wide absorption and emission bands with a strong polarization-anisotropy [1]. High doping concentrations without a significant luminescence quenching and distortions of the host lattice can also be achieved. Although the thermal conductivity of DT crystals is moderate (~ 3 W/mK), their thermo-optical properties can be improved with a so-called "athermal" crystal cut. As a result, continuous-wave (CW) and pulsed (Q-switched and mode-locked) high-efficient "bulk", thin-disk and waveguide lasers using these crystals have been reported [1].

DTs are monoclinic crystals belonging to the point group $2/m$ [2]. Then they are biaxial crystals so that they exhibit two optical axes (OAs) located in the X–Z principal plane of the dielectric frame $\{X, Y, Z\}$, when using the convention $n_x < n_y < n_z$ for the three principal refractive indices. Note that $\{X, Y, Z\}$ corresponds to the old notation $\{N_p, N_m, N_g\}$. If a light beam is propagating along one of the optical axes, its energy is spread out over a hollow cone with a distribution of polarization states [3]. This phenomenon corresponds to the internal conical refraction (CR) effect, and recently attractive properties were reported in lasers using doped with trivalent lanthanide ions DT crystals cut along this direction. For example, in Yb^{3+} :KGdW CR-laser, the tuning of the polarization state without the loss of the output power was demonstrated [4]; in Nd^{3+} :KGdW CR-laser, a unique laser mode stabilization was detected [5]. The first $2\ \mu m$ CR laser was implemented using Ho^{3+} :KYW [6].

For biaxial crystals, the orientation of the OAs can rotate as a function of the wavelength in the X–Z principal dielectric frame. This rotation can be calculated with a great precision using the dispersion equations of the three principal refractive indices. However, a real key issue is that DTs are monoclinic crystals where the orientation between the dielectric frame $\{X, Y, Z\}$ and the crystallographic frame $\{a, b, c\}$ can also rotate as a function of the wavelength λ [7]. Then both rotations have to be taken into account to reach the real orientation of the OAs and it is of particular interest at the laser wavelength λ_L when aiming to use the internal conical refraction effect in a CR-laser [8].

In the present paper, we report for the first time a complete and accurate direct measurement of the rotation of the dielectric frame $\{X, Y, Z\}$, and the values of the three principal refractive indices (n_x, n_y, n_z) over the wavelength range where CR effect can be used. This was performed in three monoclinic DTs: KGdW, KYW and KLuW to calculate the corresponding real orientation of their OAs as a function of the wavelength.

2. Rotation of the dielectric frame as a function of wavelength

In monoclinic crystals from point group $2/m$, one of the three crystallographic axes is called the "special axis" since it is linked to matter, and parallel to the two-fold axis [9]. This special axis is then perpendicular to the mirror plane m containing the two other crystallographic axes making an angle labeled β that is larger than 90° (see Fig. 1). Consequently, the crystallographic frame $\{a, b, c\}$ cannot correspond to the orthonormal dielectric frame $\{X, Y, Z\}$. According to the classical convention, b is taken as the special axis and was reported coinciding with the dielectric X-axis in DT crystals [1, 2]. So the four axes, *i.e.* a, c, Y and Z , are located in the mirror plane perpendicular to the b -axis; the angles $Z^{\wedge}c = \varphi$ and $Y^{\wedge}a = \delta$ are linked together and generally have a non-zero value.

An important feature of monoclinic crystals is that the orientation of the dielectric frame $\{X, Y, Z\}$ can rotate in the mirror plane perpendicular to the special axis as a function of any dispersive parameter of the refractive indices, as the wavelength λ for example [7]. In monoclinic DT crystals where the b -axis coincides with the dielectric X-axis, the Y–Z principal dielectric plane is concerned, its orientation as a function of λ being described by the angles $\varphi(\lambda)$ and $\delta(\lambda)$ as shown in Fig. 1 where the wavelength is equal to 0.6328 μm .

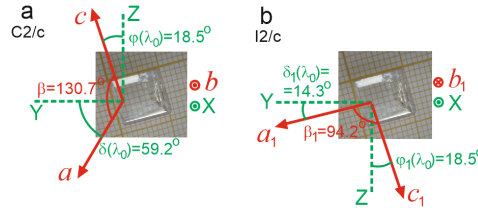


Fig. 1. $\text{Yb}^{3+}:\text{KLuW}$: Orientation of the dielectric frame $\{X, Y, Z\}$ at $\lambda_0 = 0.6328 \mu\text{m}$ and of the crystallographic frame $\{a, b, c\}$ using the crystallographic setting (a) $C2/c$, and (b) $I2/c$.

To determine the $\varphi(\lambda)$ and $\delta(\lambda)$ angles, we combined at room temperature angular data recorded from X-ray backscattered Laue geometry and conoscopy at $\lambda_0 = 0.6328 \mu\text{m}$ with a precision of 0.2° . The three DT crystals, namely KGdW, KYW and KLuW, were studied, the first one being doped with 3 at.% Nd^{3+} , the two others with 5 at.% Yb^{3+} , due to the availability of samples. According to our previous studies, the doping of DTs with the rare-earth ions in low concentrations (3-5 at.%) has a negligible effect on the orientation of the optical indicatrix. Three slabs were cut from these crystals with two faces perpendicular to the b special axis and polished to optical quality. Their dimensions were $8.28 \times 8.14 \times 4.08(b) \text{ mm}^3$, $11 \times 7.5 \times 2.77(b) \text{ mm}^3$ and $5.4 \times 5.0 \times 4.9(b) \text{ mm}^3$, respectively. We used the $C2/c$ and $I2/c$ crystallographic settings (Fig. 1) since both are possible and typically used for DT crystals, the transformation rule being $a_1 = a + c$, $b_1 = -b$ and $c_1 = -c$ and subscript 1 standing for the $I2/c$ setting. Then $Z^{\wedge}c = \varphi(\lambda) = Z^{\wedge}c_1 = \varphi_1(\lambda)$ and $Y^{\wedge}a = \delta(\lambda) \neq Y^{\wedge}a_1 = \delta_1(\lambda)$. Note that using the $C2/c$ setting, the relation between the angles $\varphi(\lambda)$ and $\delta(\lambda)$ is $\delta(\lambda) = \varphi(\lambda) + (\beta - 90^\circ)$, while it is $\delta_1(\lambda) = \varphi_1(\lambda) - (\beta_1 - 90^\circ)$ using the $I2/c$ setting. $\text{Yb}^{3+}:\text{KLuW}$ crystal is depicted in Fig. 1, showing also the relative orientation between the $\{a, b, c\}$ and $\{X, Y, Z\}$ frames at $\lambda_0 = 0.6328 \mu\text{m}$. The corresponding values of β , β_1 , $\varphi(\lambda_0)$, $\delta(\lambda_0)$ and $\delta_1(\lambda_0)$ are summarized in Table 1 for the three studied DT crystals. The values of the angles $\varphi(\lambda_0)$ and $\delta(\lambda_0)$ appear to change by less than 1° , in the row KGdW–KYW–KLuW.

Table 1. Orientation between the Crystallographic and Dielectric Frames in Three DT Crystals at $\lambda_0 = 0.6328 \mu\text{m}$ and at Room Temperature

Crystal	$C2/c$ setting			$I2/c$ setting	
	$\beta = a^{\wedge}c$	$\varphi = Z^{\wedge}c = Z^{\wedge}c_1$	$\delta = Y^{\wedge}a$	$\beta_1 = a_1^{\wedge}c_1$	$\delta_1 = Y^{\wedge}a_1$
$\text{Nd}^{3+}:\text{KGdW}$	130.8°	19.2°	60.0°	94.4°	14.8°
$\text{Yb}^{3+}:\text{KYW}$	130.7°	18.9°	59.6°	94.4°	14.5°
$\text{Yb}^{3+}:\text{KLuW}$	130.7°	18.5°	59.2°	94.2°	14.3°

We verified the orientation between the crystallographic and dielectric frames as a function of the wavelength, by placing successively the three previous slabs at normal incidence between two crossed polarizers. They were simultaneously illuminated by two parallel beams: one emitted by a He–Ne laser ($\lambda_0 = 0.6328 \mu\text{m}$), and the other one by a pulsed optical parametric oscillator (OPO) of 5-ns-FWHM pulse duration and 10 Hz repetition rate that was tunable between $\lambda = 0.4$ and $2.4 \mu\text{m}$. The slabs were rotated around the b -axis using a piezoelectric motor [7]. The transmitted power follows Malus law, the positions of minima providing a direct access to the angle $\varphi(\lambda) = Z^{\wedge}c$ that was recorded between 0.4 and $1.6 \mu\text{m}$ with a precision of 0.2° .

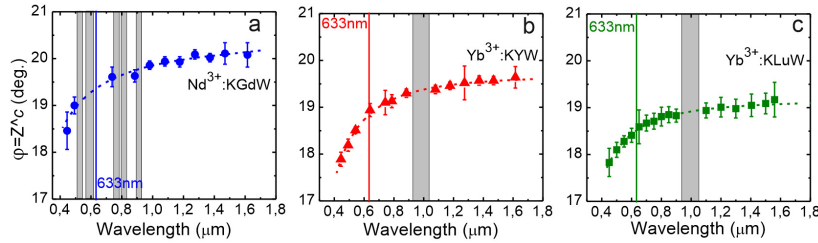


Fig. 2. $\varphi(\lambda)$ in (a) $\text{Nd}^{3+}:\text{KGdW}$, (b) $\text{Yb}^{3+}:\text{KYW}$, and (c) $\text{Yb}^{3+}:\text{KLuW}$ crystals; *dots* are our data, *grey regions* stand for the absorption ranges of Nd^{3+} and Yb^{3+} ions and vertical lines for $\lambda_0 = 0.6328 \mu\text{m}$, *dashed curves* are the fittings of the experimental data with Eq. (1).

The behavior of $\varphi(\lambda)$ is shown in Fig. 2 where the vertical lines mark out the angle $\varphi(\lambda_0 = 0.6328 \mu\text{m})$ determined using conoscopy, while the grey regions correspond to the Yb^{3+} or Nd^{3+} ions absorption ranges. It appears similar in the three DT crystals and is not affected by the lanthanide ions absorption ranges: it increases continuously with λ , its value changing more rapidly in the visible range (0.4–0.8 μm) than in the near-IR one. Its overall variation is equal to about 2° between 0.4 and 1.6 μm . The experimental points in Fig. 2 were fitted with the following dispersion formula:

$$\varphi(\lambda) = A + \frac{B}{\lambda} + \frac{C}{\lambda^2}, \quad (1)$$

and the best-fit A , B and C coefficients are listed in Table 2, λ being expressed in μm .

Table 2. Coefficients at Room Temperature Representing The Rotation of the Dielectric Frame* of DT Crystals

Crystal	A	$B, \mu\text{m}$	$C, \mu\text{m}^{-2}$
$\text{Nd}^{3+}:\text{KGdW}$	20.51	−0.5364	−0.1205
$\text{Yb}^{3+}:\text{KYW}$	19.67	+ 0.1135	−0.4047
$\text{Yb}^{3+}:\text{KLuW}$	19.15	+ 0.0712	−0.3001

*Coefficients of the formula for the $\varphi(\lambda) = Z^*c$ angle, see Eq. (1).

3. Dispersion equations for the three principal refractive indices

The three principal refractive indices, (n_x, n_y, n_z) , were determined by using the minimum deviation method. The details about the use of this method for biaxial crystals can be found in [10]. The propagation of light in the principal planes of the biaxial crystals is described in details in [11]. Undoped KGdW, KYW and KLuW crystals were cut each as a triangular prism of cm-size, with the apex angle ranging between 25 and 30° for each prism. During the prism cutting, the laser alignment was used resulting in the precision of the orientation of $\sim 1'$. The prisms polished to optical quality were placed successively at the center of a goniometer providing angular measurements with a precision of $2''$. They were combined to a He-Ne laser, a halogen lamp, a set of laser diodes, and a wavelength-tunable OPO. These light sources gave access to several wavelengths distributed over the spectral range of 0.4–1.6 μm . The light polarization state was established and controlled using Glan-Taylor polarizers.

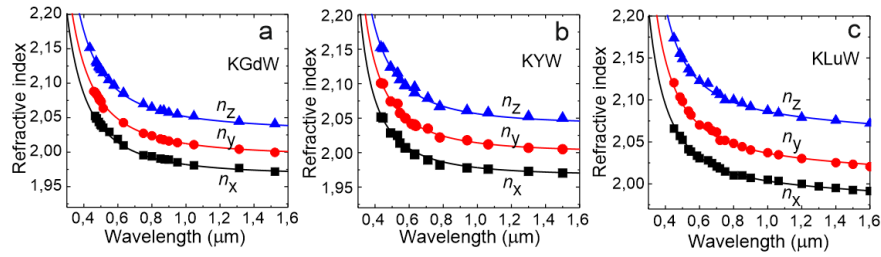


Fig. 3. Variation of the three principal refractive indices, n_x , n_y and n_z as a function of the wavelength in (a) KGdW, (b) KYW and (c) KLuW crystals; dots are our experimental data, continuous lines are Eq. (1) with parameters of Table 3.

Using the appropriate polarization scheme along each dielectric axis of the oriented prisms and our setup, the wavelength dispersion of the three principal refractive indices, (n_x , n_y , n_z), were determined in undoped KGdW, KYW and KLuW crystals with a precision of 2×10^{-4} . They are shown in Fig. 3 with their simultaneous fit by using the Levenberg-Marquardt algorithm, and using the same following form of Sellmeier equation with an infrared correction:

$$n_i^2(\lambda) = A_i + \frac{B_i \lambda^2}{\lambda^2 - C_i^2} - D_i \lambda^2, \quad (2)$$

where A_i , B_i , C_i and D_i ($i = x, y$ or z) are the fitting parameters, λ being expressed in μm . The corresponding best result is shown in Fig. 3(a)-3(c) up to 1.6 μm . It was established with the final values of the fitting parameters summarized in Table 3. Note that we refined the Sellmeier equations of KLuW from [1].

Table 3. Sellmeier Coefficients* at Room Temperature for the Principal Refractive Indices of DT Crystals

Crystal	n_i	A_i	B_i	$C_i, \mu\text{m}$	$D_i, \mu\text{m}^{-2}$
KGdW	n_x	0.9927	2.8661	0.1523	-0.00139
	n_y	0.8753	3.0976	0.1554	-0.00042
	n_z	1.5157	2.6141	0.1733	+0.00183
KYW	n_x	2.6986	1.1578	0.2127	-0.00215
	n_y	2.5253	1.4638	0.2083	-0.00237
	n_z	2.3951	1.7564	0.2025	-0.00226
KLuW	n_x	2.1777	1.7870	0.1751	+0.00744
	n_y	2.2893	1.7992	0.1900	+0.00822
	n_z	2.9957	1.2978	0.2250	+0.01099

*In accordance with Eq. (2).

4. Location of the optical axes as a function of wavelength

The orientation between the two optical axes OAs and Z-axis in the X-Z principal plane of a biaxial crystal as a function of the wavelength is described by the angle $V_Z(\lambda) = \angle \text{Z}^{\circ}\text{OA}$. It can be calculated without any approximation, from the wavelength dispersion of the three principal refractive indices and the following formula [3, 9]:

$$\sin^2 V_Z = \frac{1/n_y^2 - 1/n_x^2}{1/n_z^2 - 1/n_x^2}. \quad (3)$$

Similarly, the angle of the internal conical refraction cone, $A_{\text{CR}}(\lambda)$ that corresponds to the spatial walk-off angle in this direction, is defined by:

$$\tan^2 A_{\text{CR}} = \frac{n_y^2 - n_x^2}{n_x^2} \cdot \frac{n_z^2 - n_y^2}{n_z^2}. \quad (4)$$

The calculated dispersion of angles $V_Z(\lambda)$ and $A_{CR}(\lambda)$ for KGdW, KYW and KLuW is presented in Fig. 4; the dispersion formulas for the refractive indices (n_x , n_y , n_z) are according to Eq. (2) and Table 3. The accuracy is of 5' over the wavelength range where they are valid, *i.e.* between 0.5 μm and 1.6 μm .

The value of the angle $V_Z(\lambda)$ can also be determined directly when rotating one sample cut as a sphere from the direct observation on a screen of the CR effect resulting in a double hollow cone [3, 10]. A rod cut along an OA can be also used, provided a magnification with focal lenses for better observation of the CR effect [8]. Using this last method direct measurements of $V_Z(\lambda)$ were reported in $\text{Nd}^{3+}:\text{KGdW}$ and $\text{Ho}^{3+}:\text{KYW}$ but as mentioned in [8], there was a very strong discrepancy of the order of 5° between experimental data and calculations using the Sellmeier equations from [1].

As shown in Fig. 4(a), our calculated values of $V_Z(\lambda)$ in the X-Z principal dielectric plane using our Sellmeier coefficients of KGdW and KYW crystals, corroborate measurements of [7] recorded independently. For example, both approaches show that at the laser emission wavelength $\lambda_L \sim 1 \mu\text{m}$, V_Z decreases from $43^\circ 42'$ to $40^\circ 09'$ in the series KYW–KGdW–KLuW, and A_{CR} is close between $\text{Yb}^{3+}:\text{KLuW}$ ($2^\circ 16'$) and $\text{Nb}^{3+}:\text{KGdW}$ ($2^\circ 13'$), these values being higher than in $\text{Yb}^{3+}:\text{KYW}$ ($1^\circ 57'$). Data of [7] being performed in doped DT crystals, such a good agreement can be explained by their very low doping rate.

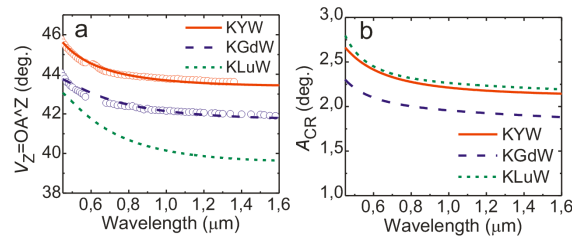


Fig. 4. Calculated values in DT crystals performed as a function of wavelength and using Eqs. (2) to (4) and Table 3 for: (a) the optical axis angle V_Z , (b) the angle of a cone of internal conical refraction A_{CR} ; symbols in (a) are experimental data from [8].

We calculated the orientation of OAs in the dielectric frame as a function of the wavelength by combining the two angles $V_Z(\lambda)$ and $\varphi(\lambda)$ that report their simultaneous rotation in two different principal dielectric planes: the angle $V_Z(\lambda)$ stands for the OA rotation in the X–Z principal plane; the angle $\varphi(\lambda)$ is due to a rotation of the Y–Z principal dielectric plane in the mirror plane that implies the same rotation of the OAs out of this plane. It shows that in monoclinic DT crystals the optical axes undergo a 3D-rotation as a function of the wavelength with respect to the crystallographic frame, as depicted in inset of Fig. 5(a). This rotation is a consequence of the rotation of the Y and Z axes, the OAs lying in the X–Z plane of the crystal. The overall variation of the angles $V_Z(\lambda)$ and $\varphi(\lambda)$ is of 2.5° between 0.45 and 1.6 μm in KGdW, KYW and KLuW crystals, see Fig. 5.

Note that using the thermo-optic coefficients of DT crystals reported so far [12], we also found that the first derivative of V_Z according to the temperature T , *i.e.* dV_Z/dT , exhibits a value of 0.2 arcmin/K in these crystals. Thus for an upper estimate of the temperature rise induced in the diode-pumped laser crystal of 200 K would lead to a change of the OAs orientation on the order of 1° . Since it has been reported that if the optical axes move by 0.5° , it is enough to violate the CR effect [2], such an additional movement of OAs can be detrimental when the use of the internal conical refraction effect is targeted for the design of a CR-laser.

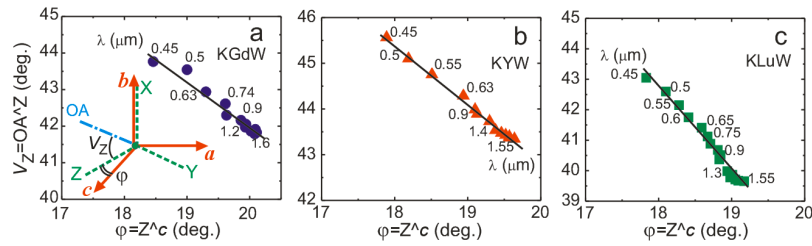


Fig. 5. Values of angles $\phi(\lambda)$ versus $V_Z(\lambda)$ in (a) KGW, (b) KYW, and (c) KLuW crystals. *Inset* of (a) is the corresponding 3D-rotation of the OA. Data close to dots are the corresponding values of λ .

5. Conclusion

We report a 3D rotation of the optical axes as a function of the wavelength, in the three monoclinic double tungstate crystals $\text{KRE}(\text{WO}_4)_2$ with $\text{RE} = \text{Gd}, \text{Y}$ or Lu . We used the direct measurement of the rotation of the dielectric frame, and the determination of the Sellmeier equations. Taking into account these data, we were able to calculate, for the first time to our knowledge, the location of the optical axes as a function of the wavelength. These data are of prime importance for the design of a laser using DT crystals cut along one optical axis.