Elasto-optic coefficients of $KGd(WO_4)_2$ crystals

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Abstract. The modules of ten components of the elasto-optic tensor for $KGd(WO_4)_2$ crystals have been determined using a Dixon–Cohen method. We have found that some of the elasto-optic components of these crystals reach notably high values. Being combined with the fact of high acoustic-wave velocity, this should imply short response times of the acousto-optic devices based on $KGd(WO_4)_2$, provided that the acousto-optic figure of merit remains as high as that calculated in the present work, $(32.6\pm4.5)\times10^{-15}$ s^3/kg.

Keywords: $KGd(WO_4)_2$ crystals, elasto-optic coefficients, acousto-optics

UDC: 535.551+535.421

1. Introduction

K*Re*$(WO_4)_2$ crystals, with *Re* being rare-earth elements such as Gd, Yb, Er, Tb, La, etc., are well known materials for the active media of solid-state lasers [1]. Besides, these crystals can be used for Raman lasers with both up and down frequency conversions [2, 3]. Among K*Re*$(WO_4)_2$ crystals, potassium-gadolinium tungstate $KGd(WO_4)_2$ (abbreviated as KGW further on) is one of the best studied materials. Since the ion radii of Gd^{3+} and Nd^{3+} are close, the KGW crystals can be easy doped, e.g., with Nd^{3+} ions [4, 5]. The crystals have high laser-damage threshold (10–20 J/cm^2 [6]), high third-order nonlinear coefficients, and low Raman conversion threshold [7]. Finally, KGW is optically transparent from 0.3 to 5 μm [6].

The KGW crystals belong to the monoclinic system under normal conditions. They are characterized by the point symmetry group 2/m and the space group $C2/c$. The unit cell parameters are equal to a = 10.652 Å, b = 10.374 Å, c = 7.582 Å and β = 130.80 deg [8]. Notice that the unit cell parameters could be equal to a' = 7.582 Å, b' = 10.374 Å, c' = 8.087 Å and β' = 94.41 deg if the space symmetry group $I2/a$ were assumed. Although the last fact contradicts the common recommendations of setting [9], the habitus of the crystal corresponds to just this choice of the unit cell. To transform crystallographic coordinate system to Cartesian crystal-physical one, we use the crystallographic setting [10], where a''= 8.087 Å, b'' = 10.374 Å, c'' = 7.582 Å and β″= 94.41 deg (see Fig. 1). Then the monoclinic angle becomes closer to 90 deg and our results still can be comparable with those reported in Refs. [10–12].

KGW is optically biaxial, with the angle V = 43.3 deg between the optic axes at the light wavelength λ = 632.8 nm [13]. The principal axis N_p of optical indicatrix coincides with the principal crystallographic axis b, while the N_g axis is rotated clockwise (if one looks along the b'' axis) by the angle φ = 21.5 deg with respect to the c'' axis in the $a''c''$ plane [8, 10].

Recently it has been shown that the KGW crystals can be efficiently utilized in various acousto-optic (AO) devices, e.g. modulators or collinear filters [10, 11]. Such applications expect some knowledge of elasto-optic (EO) coefficients p_{ij} because it is these coefficients that determine the AO figure of merit M_2 of a crystal. The appropriate relationship is $M_2 = n^6 p_{eff}^2 / \rho v^3$,

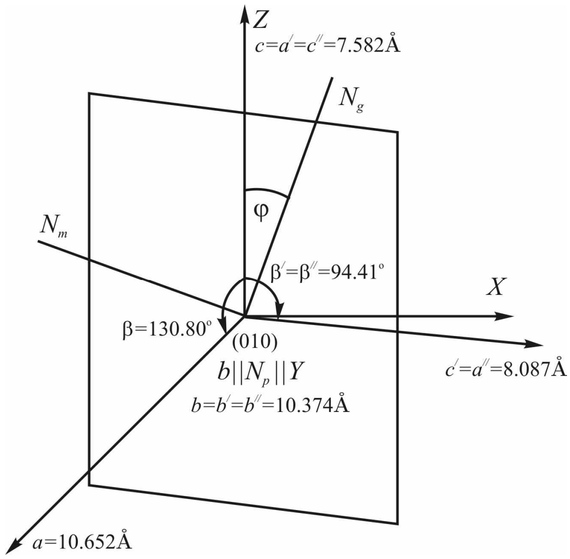

Fig. 1. Crystallographic (*abc*, β ; $a'b'c',\beta'$; $a''b''c'',\beta''$), crystal-physical (*XYZ*) and crystal-optical ($N_pN_mN_g$) settings adopted for the KGW crystals.

with n being the refractive indices that corresponds to the polarizations of interacting optical waves, p_{eff} the effective EO coefficient, v the acoustic-wave (AW) velocity, and ρ the crystal density.

In fact, twelve components of the EO tensor have been determined in Refs. [10, 12]. Let us remind that the EO tensor for the point symmetry group 2/m contains twenty components, nineteen of which are invariants [14]. Besides, EO components have been determined in Refs. [10, 12] in a so-called crystal-optical coordinate system (see Fig. 1). This is the system associated with the eigenvectors of optical-frequency impermeability tensor $B_k = \left(1/n^2\right)_k$. The system is very sensitive to external actions and even to scalar ones. Namely, any change in the temperature, hydrostatic pressure or the wavelength of optical radiation would lead to rotation of optical indicatrix around the two-fold symmetry axis, and so to the rotation of crystal-optical coordinate system. Moreover, the coordinate eigensystems for different tensorial properties would differ from the crystal-optical one. This is the reason why determination of tensorial constitutive parameters of low symmetry crystals is more reasonable to carry out in crystal-physical Cartesian coordinate system *XYZ*, which is associated with the crystallographic one $a''b''c''$ as shown in Fig. 1.

In the present work we aim at studying the EO parameters of KGW in its crystal-physical coordinate system.

2. Experimental procedures and phenomenological relations

The EO properties of KGW were measured in the crystal-physical Cartesian coordinate system *XYZ* linked to the crystallographic system $a''b''c''$ as illustrated in Fig. 1. The relations among the appropriate axes are as follows: $Z \parallel c''$, $Y \parallel b'' \parallel N_p$ and $X \perp b''c''$. The EO properties were studied using a standard Dixon–Cohen method [15]. It is based on comparison of AO parameters of a material under study with those of a standard material. In our case we used fused silica as a standard material. The scheme of our experimental setup was described elsewhere (see Ref. [16]). The KGW crystals were supplied by the “Plant “Optika” Company (Republic of Belarus).

A sample under test was prepared in the shape of cube with the sizes ~ $1\times1\times1$ cm^3 and the faces perpendicular to the crystal-physical axes. The test sample was bonded with the reference sample made of a standard material. The longitudinal AW in the cell was excited with a piezoelectric transducer fabricated from $LiNbO_3$ crystals. The excited wave with the frequency $f = 50$ MHz was modulated by square pulses with the duration 0.5 μs and the repetition rate 300 μs.

The AO figure of merit of a sample under study can be calculated as

$$M_2^{sa} = M_2^{st} \frac{I_{st}}{I_{sa}} \left(\frac{I_3 I_4}{I_1 I_5} \right)^{1/2} , \quad (1)$$

where $M_2^{st} = 1.56\times10^{-15}$ s^3/kg is the AO figure of merit of fused silica in the case of light diffraction at the longitudinal AW [1]. In Eq. (1), I_{sa} and I_{st} are the intensities of light ($\lambda = 632.8$ nm) transmitted respectively through the test and standard samples, which have been detected with no acoustic signal applied. The intensities of diffracted light that correspond to AO interactions with the forward-propagating acoustic pulse for the cases of standard material and test sample are referred to as I_1 and I_3. Finally, the intensities of the pulse reflected from the rear surface of the sample and the same pulse re-entered in the standard sample are denoted as I_4 and I_5, respectively.

The effective EO coefficient can be calculated using the relation [17]

$$\left| p_{eff} \right| = \frac{\sqrt{M_2^{sa} \rho v^3}}{n^3} , \quad (2)$$

where $\rho = 7216$ kg/m^3 is the density of KGW. Here the AW velocities have been calculated from the elastic stiffness coefficients [10] by solving a Christoffel equation.

The refractive indices for the optical waves polarized parallel to the principal axes of the crystal-physical coordinate system XYZ were calculated using the formulae

$$n_X = \left(\frac{\cos^2\varphi}{N_m^2} + \frac{\sin^2\varphi}{N_g^2} \right)^{-1/2} , \; n_Z = \left(\frac{\sin^2\varphi}{N_m^2} + \frac{\cos^2\varphi}{N_g^2} \right)^{-1/2} , \; n_Y = N_p . \quad (3)$$

As mentioned above, the angle φ is equal to $\varphi = 21.5$ deg according to Ref. [8]. Nonetheless, we have re-measured this angle using the method of observing conoscopic patterns at $\lambda = 632.8$ nm. One of the faces of our sample is perpendicular to the c'' axis, which has been verified with an X-ray analysis. We have found that rotation of our sample around the b'' axis leads to appearance of conoscopic pattern typical for the direction of principal axis of the optical indicatrix. In this manner we have determined the angle between the direction N_g and the c'' axis, which equals to $\varphi = 14.5$ deg.

In order to determine the EO coefficients, one has to consider all the possible types of isotropic AO interactions. We will solve this problem step by step. Let the incident optical wave with the electrical induction D_l that propagates along the Z axis and is polarized parallel to the Y axis interact with the longitudinal AW propagating along the X axis. This corresponds to AO interactions of a so-called type 1 (see Fig. 2a). Then the relation for the electric field of diffracted optical wave is given by $E_2 = \Delta B_2 D_2 = p_{21} e_{12}$ (with e_j being the strain component caused by the AW). The appropriate effective EO coefficient can be written as $p_{eff} = p_{21}$.

When the incident optical wave propagating along the Z axis with its polarization parallel to the X axis interacts with the longitudinal AW propagating along the X axis (the AO interactions of the type 2 – see Fig. 2a), one can determine the effective EO coefficient $p_{eff} = p_{11}$ basing on the relation $E_1 = \Delta B_1 D_1 = p_{11} e_1 D_1$. Let the incident light wave polarized parallel to the Y axis propagate along the X axis and interact with the longitudinal AW propagating along the Z axis (the type 3 – see Fig. 2a). In this case we obtain $E_2 = \Delta B_2 D_2 = p_{23} e_3 D_2$ and $p_{eff} = p_{23}$. When the incident light wave polarized parallel to the Z axis propagates in the XZ plane along the X axis, whereas the longitudinal AW propagates along the Z axis, one deals with the AO interaction type 4 (see Fig. 2a). Then the relations $E_3 = p_{33} e_3 D_3$ and $p_{eff} = p_{33}$ hold true. Moreover, the EO coefficient remains the same ($p_{eff} = p_{33}$) in the case of AO interactions in the YZ plane, when the incident optical wave propagates along the Y axis and is polarized along the Z axis.

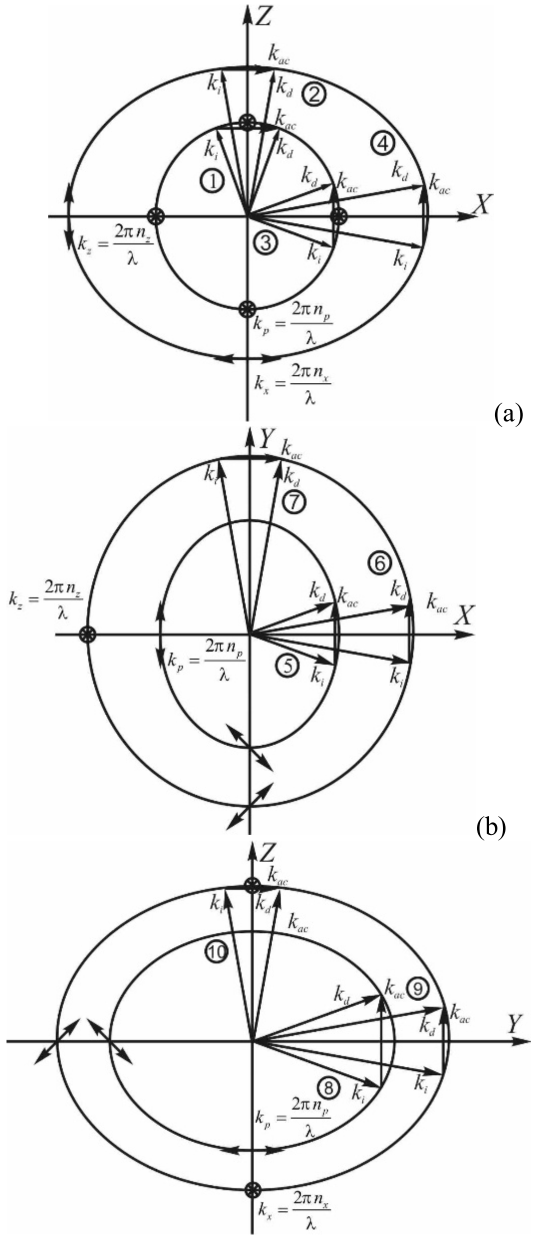

Fig. 2. Schematic vector diagrams for AO interactions in KGW in the coordinate planes *XZ* (a), *XY* (b) and *YZ* (c): k_i, k_d and k_{ac} are wave vectors of the incident optical wave, diffracted optical wave and the AW, respectively. Double-sided arrows and crossed circles indicate polarizations of optical eigenwaves.

Let the incident light wave polarized parallel to the Y axis propagate in the XY plane along the X axis and interact with the longitudinal AW propagating in the same plane along the Y axis (the AO interactions of the type 5 – see Fig. 2b). Then one can write out the constitutive relation as $E_2 = \Delta B_2 D_2 = p_{22} e_2 D_2$, while the EO coefficient is given by $p_{eff} = p_{22}$. When the incident light wave polarized parallel to the Z axis propagates along the X axis and interacts with the longitudinal AW propagating along the Y axis (the case of AO interactions of the type 6 – see Fig. 2b), the AO relationships are as follows: $E_3 = \Delta B_3 D_3$, $\Delta B_3 = p_{32} e_2$, and $E_3 = p_{32} e_2 D_2$. When the incident optical wave propagates along the Y axis and is polarized along the Z axis (the type 7 – see Fig. 2b), the formulae describing AO interactions of this wave with the longitudinal AW propagating along the X axis can be written as $E_3 = \Delta B_3 D_3 = p_{31} e_1 D_1$ and $p_{eff} = p_{31}$.

Let the optical wave polarized parallel to the Z axis propagate along the Y axis. Its AO interactions with the transverse AW that propagates along the Z axis and is polarized parallel to the X axis (the interactions of the type 8 – see Fig. 2c) can be described by the formula $E_3 = 0.5 p_{35} e_5 D_3$, while the effective EO coefficient is defined as $p_{eff} = p_{35}$. This effective coefficient can also be measured in the case of AO interactions in the XY plane, when the optical wave polarized along the Z axis propagates along the Y axis and interacts with the transverse AW propagating along the X axis and polarized along the Z axis (the type 7 – see Fig. 2b). When the incident light wave with the polarization parallel to the X axis propagates along the Y axis and interacts with the longitudinal AW propagating along the Z axis (the type 9 – see Fig. 2c), the AO relationships can be written in the form $E_1 = p_{13} e_3 D_1$ and $p_{eff} = p_{13}$. Finally, the effective EO coefficient $p_{eff} = p_{12}$ is actual at the geometry when the incident light wave polarized parallel to the X axis propagates along the Z axis and interacts with the longitudinal AW propagating along the Y axis (the type 10 – see Fig. 2c). Then we obtain the formulae $E_1 = \Delta B_1 D_1 = p_{12} e_2 D_1$ and $p_{eff} = p_{12}$.

3. Results and discussion

As mentioned above, the angle φ determined in the present work has proved to be almost twice as smaller as the value reported in Ref. [8]. This angle follows from the relation $\tan 2\varphi = 2B_5 / (B_1 - B_3)$, where B_k are the components of optical impermeability tensor written in the crystallographic coordinate system. The disagreement between the corresponding φ values can be caused by the changes observed for the refractive indices N_m and N_g. In fact, a small variation in the third digit after decimal point for the refractive indices is quite enough in order for the φ angle to change from 21.5 to 13.5 deg. Notice that different literature data [8, 18] report somewhat different refractive indices of the KGW crystals (see Table 1). This fact can lead to changing angle between the optic axes [13] and changing rotation angle of optical indicatrix with respect to the crystal-physical coordinate system. Taking into account the fact that the principal refractive indices for KGW reported in Refs. [8, 18] are very similar and differ only in the second digit after decimal point, we accept $N_g = 2.09$, $N_p = 2.01$ and $N_m = 2.04$ in our further calculations.

Table 1. Refractive indices for the KGW crystals at $\lambda = 632.8$ nm .

Reference	N_g	N_m	N_p
[8]	2.0950	2.0414	2.0116
[18]	2.08608	2.04580	2.01348

The modules of the ten EO components determined by us are gathered in Table 2. The angle of non-orthogonality of the polarizations of AWs does not exceed ~ 10 deg whenever the AWs propagate along the *X*, *Y* or *Z* axes. As a consequence, one can neglect this angle at this stage of consideration. Then the AWs can be regarded as purely longitudinal and transverse. In this case the modules of the effective EO coefficients determined above would correspond to the modules of single components of the EO tensor.

Table 2. Geometries of AO interactions analyzed and EO coefficients found for the KGW crystals.

Type of AO interaction	Directions of AW propagation and polarization, and AW velocity (in m/s)	Directions of light propagation and polarization	Refractive index *n*	AO figure of merit M_2, 10^{-15} s^3/kg (in the crystal-physical coordinate system)	Effective EO coefficient	Module of EO coefficient (in the crystal-physical coordinate system)	EO coefficient according to Ref. [12] (in the crystal-optical coordinate system)
1	*X*, *X*, and 5062	*Z*, *Y*	$n_Y = 2.01$	0.8±0.2	p_{21}	0.11	0.13
2	*X*, *X*, and 5062	*Z*, *X*	$n_X = 2.04$	2.4±0.1	p_{11}	0.18	0.11
3	*Z*, *Z*, and 4334	*X*, *Y*	$n_Y = 2.01$	4.6±1.0	p_{23}	0.21	0.23
4	*Z*, *Z*, and 4334	*X*, *Z*	$n_Z = 2.09$	32.6±4.5	p_{33}	0.49	0.28
5	*Y*, *Y*, and 4878	*X*, *Y*	$n_Y = 2.01$	0.52±0.04	p_{22}	0.08	0.04
6	*Y*, *Y*, and 4878	*X*, *Z*	$n_Z = 2.09$	0.50±0.04	p_{32}	0.07	0.09
7	*X*, *X*, and 5062	*Y*, *Z*	$n_Z = 2.09$	0.5±0.2	p_{31}	0.07	0.13
8	*Z*, *X*, and 2402	*Y*, *Z*	$n_Z = 2.09$	1.51±0.04	p_{35}	0.04	–0.13
9	*Z*, *Z*, and 4334	*Y*, *X*	$n_X = 2.04$	4.0±0.2	p_{13}	0.18	0.23
10	*Y*, *Y*, and 4878	*Z*, *X*	$n_X = 2.04$	1.3±0.1	p_{12}	0.12	0.14

Although the angle φ is not very large, some of the EO coefficients determined in the present work and in Ref. [12] differ conspicuously. In particular, this concerns the tensor components p_{11}, p_{33} and p_{31}. It is these coefficients that change their values when the EO tensor is rewritten from the crystal-optical coordinate system to the crystal-physical one. According to our measurements, the coefficient p_{22}, which is invariant with respect to the rotations of optical indicatrix around the b'' axis, is equal to 0.08 (to be compared with the value 0.04 reported in Ref. [12]. Due to our data, the corresponding AO figure of merit proves to be almost five times higher than that following from the data [12]. Notice that the AW propagating along the two-fold axis is purely longitudinal. Hence, the non-orthogonality of its polarization is equal to zero and so cannot affect the p_{eff} value. The coefficient p_{35} differs essentially, too. It is known that this coefficient is sensitive to the rotation of crystal-optical coordinate system with respect to the crystal-physical system.

As seen from Table 2, the maximal AO figure of merit, $(32.6\pm4.5)\times10^{-15}$ s^3/kg, is achieved for the interaction geometry #4, due to high EO coefficient. This value is almost the same as the maximal value reported in Ref. [12], which is reached in the case of AO interactions with the quasi-transverse wave. This fact implies that the KGW crystals can be characterized by higher AO figures of merit in the case when the optical waves interact with the AWs propagating in some directions, which do not coincide with the principal axes of the coordinate system. Nevertheless, one should know the values of all EO tensor components in order to analyze the anisotropy of AO figure of merit (see, e.g., Ref. [19]). This will be a subject of our further studies.

4. Conclusion

Using the AO Dixon–Cohen method, we have determined the modules of ten components of the EO tensor for the KGW crystals in its crystal-physical coordinate system. It has been found that some of the EO tensor components of KGW reach high enough values. In particular, the component p_{33} is equal to 0.49. This fact, together with high enough AW velocity (4334 m/s) would lead to short response times of the AO devices based on KGW, whenever the AO figure of merit remains high enough, e.g. $(32.6\pm4.5)\times10^{-15}$ s^3/kg as found above. We have also observed that such important parameters of the KGW crystals as refractive indices can vary notably for the samples of different origins. Finally, we have demonstrated that the optical indicatrix in our crystals rotates by the angle 14.5 deg with respect to the crystallographic axis, although this angle has been accepted to be 21.5 deg in the literature.

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Анотація. *Методом Діксона-Коена в роботі визначено модулі десяти компонент пружно-оптичного тензора кристалів $KGd(WO_4)_2$. Виявлено, що деякі з компонент досягають високого значення, що разом з високими швидкостями поширення акустичних хвиль може привести до значної швидкодії акусто-оптичних пристроїв на основі кристалів $KGd(WO_4)_2$ при порівняно великих значеннях коефіцієнта акусто-оптичної якості ($(32.6\pm4.5)\times10^{-15}$ с³/кг).*