Anisotropy of acousto-optic figure of merit in tetragonal crystals with accounting for non-orthogonality of acoustic eigenwave polarizations.

# 1. The cases of KH<sub>2</sub>PO<sub>4</sub> and NH<sub>4</sub>H<sub>2</sub>PO<sub>4</sub> crystals

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**Abstract.** In the present work we derive analytical relations for the effective elastooptic coefficients with accounting for the non-orthogonality of polarizations of the acoustic waves (AWs) in the crystals that belong to the symmetry groups  $\overline{4}2m$ , 4/mmm, 422 and 4mm. We analyze anisotropies of acousto-optic (AO) figure of merit (AOFM) for KH<sub>2</sub>PO<sub>4</sub> and NH<sub>4</sub>H<sub>2</sub>PO<sub>4</sub> crystals within their crystallographic planes, which are derived under conditions when the AW non-orthogonality effect is neglected by or accounted for. We find that consideration of the angle of AW nonorthogonality changes significantly the AOFM values. AO diffraction geometries corresponding to maximums of AOFM are obtained for all of the nine types of AO interactions, including those concerned with the collinear diffraction. We demonstrate that the principal AOFM maximum for NH<sub>4</sub>H<sub>2</sub>PO<sub>4</sub> is equal to  $10.0 \times 10^{-15}$  s<sup>3</sup>/kg. It can be achieved at the type II of AO interactions with the quasilongitudinal AW in the crystallographic plane ab. The highest AOFM value peculiar for the anisotropic AO diffraction in  $NH_4H_2PO_4$ ,  $9.5\times10^{-15}\,\text{s}^3/\text{kg}$ , can be reached at the type IX of AO interactions with a so-called AW QT<sub>2</sub> in the ac plane. Finally, the highest AOFM found for the case of collinear diffraction,  $4.4 \times 10^{-15}$  s<sup>3</sup>/kg, is also inherent to the NH<sub>4</sub>H<sub>2</sub>PO<sub>4</sub> crystals, KH<sub>2</sub>PO<sub>4</sub> is characterized by somewhat lower AOFM values.

**Keywords:** acousto-optic figure of merit, anisotropy, tetragonal crystals, KH<sub>2</sub>PO<sub>4</sub>, NH<sub>4</sub>H<sub>2</sub>PO<sub>4</sub>, non-orthogonality of acoustic wave polarization

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# 1. Introduction

A great number of efficient acousto-optic (AO) materials belong to the crystals of tetragonal system. One of these crystalline materials is paratellurite (the chemical formula  $TeO_2$  and the point symmetry group 422 [1]), of which AO figure of merit (AOFM) is as high as  $1200 \times 10^{-15} \text{s}^3/\text{kg}$  [2]. We remind that the AOFM is determined by the relationship  $M_2 = n^6 p_{ef}^2 / \rho v^3$ , where  $\rho$  denotes the material density, n the refractive index,  $p_{ef}$  the effective elasto-optic coefficient (EEC), and v the velocity of the acoustic wave (AW). One of the crystals representing tetragonal AO materials is lithium tetraborate,  $\text{Li}_2\text{B}_4\text{O}_7$  which is described by the point symmetry group 4mm. It is highly resistant to a high-power optical radiation, with the appropriate damage threshold being equal to  $32.2 \text{ GW/cm}^2$  (according to our measurements [3]) or even  $40 \text{ GW/cm}^2$  (according to the data of Ref. [4]). The above crystals are transparent in the deep ultraviolet spectral range down to 170 nm [4]. Hence, lithium tetraborate represents a promising AO material [5–8] for operating high-power

optical radiation in the ultraviolet range. Following from the results of our study [8], its maximal AOFM can reach  $3.44 \times 10^{-15}$  s<sup>3</sup>/kg in the visible spectral range. In principle, it can become still higher in the ultraviolet range due to dispersion of refractive indices and elasto-optic coefficients.

 $\overline{4}$ 2m under normal conditions also seem to be efficient AO materials for the ultraviolet range and high-power optical radiation. These crystals are transparent down to 250 and 200 nm, respectively [1]. As a result, their AO properties have attracted much attention associated with ultraviolet tunable AO filters [9–12]. As shown by the present authors, the maximal AOFM value for the KDP crystals is equal to  $7.1\times10^{-15}\,\text{s}^3/\text{kg}$  [13]. According to the literature data, the AOFM for the ADP crystals amounts to  $6.43\times10^{-15}\,\text{s}^3/\text{kg}$  [1]. However, the last values do not represent the maximal AOFMs possible for the above crystals.

In our previous works, we have developed an analytical method for the studies of AOFM anisotropy [8, 13-19]. Among the crystals of which AOFMs have been analyzed there are such tetragonal crystals as KDP, TeO<sub>2</sub>, Li<sub>2</sub>B<sub>4</sub>O<sub>7</sub> and NaBi(MoO<sub>4</sub>)<sub>2</sub>. As a matter of fact, this analysis has been carried out under assumption that the acoustic eigenwaves are purely transverse or longitudinal. This approximation is valid only when the angle of polarization deviation from the purely longitudinal or transverse states is very small (see, e.g., Mys O et. al, 2018. Ukr. J. Phys. Opt. 19: 165). As shown in Ref. [20] for the alternative case, consideration of the deviation of AW polarizations from the purely longitudinal or transverse states changes essentially the relations for the EEC and, as result, affects the final AOFM values. As follows from the results [20], the analytical relations for the AOFM with accounting for the AW non-orthogonality can be obtained only under simplest conditions when the AWs propagate within the principal crystallographic planes. Otherwise, the Christoffell tensor contains more than one off-diagonal components and can be expressed in the coordinate eigensystem using numerical solutions only. Nonetheless, even the correct results obtained for the principal crystallographic planes would provide important possibilities to design AO devices with the maximal AOFM values. Our aim is to analyze the AOFM anisotropy in a number of tetragonal crystals such as KDP, TeO<sub>2</sub>, Li<sub>2</sub>B<sub>4</sub>O<sub>7</sub> and NaBi(MoO<sub>4</sub>)<sub>2</sub>, while accounting properly for the AW non-orthogonality. Notice that the AOFM anisotropy in the ADP crystals is studied here for the first time. In the first part of this work, we report the results obtained for the KDP and ADP crystals. The second part of the present study will deal with the other tetragonal crystals mentioned above.

## 2. Methods of analysis

# 2.1. Acoustic and optical properties of KDP and ADP crystals

We remind the main properties of the KDP and ADP crystals, which are needed for our further analysis. KDP and ADP are optically uniaxial and negative at the room temperature. At the light wavelength  $\lambda=632.8$  nm , the refractive indices are equal to  $n_o=1.5073$  and  $n_e=1.4668$  for KDP [21], and  $n_o=1.53$  and  $n_e=1.48$  [22] for ADP. The densities of the KDP and ADP crystals are equal to  $\rho=2338$  kg/m³ and  $\rho=1803$  kg/m³, respectively [1]. In our calculations, we use the elastic-stiffness coefficients of the KDP crystals obtained in Ref. [23] ( $C_{11}=71.4\pm0.8$ ,  $C_{12}=-4.9\pm1.0$ ,  $C_{13}=12.9\pm0.3$ ,  $C_{33}=56.15\pm0.3$ ,  $C_{44}=12.7\pm0.1$  and  $C_{66}=6.24\pm0.05$  GPa) and the elasto-optic coefficients determined in Ref. [24] ( $p_{11}=0.238\pm0.024$ ,  $p_{12}=0.249\pm0.013$ ,  $p_{13}=0.242\pm0.012$ ,  $p_{31}=0.227\pm0.011$ ,  $p_{33}=0.242\pm0.024$ ,  $p_{44}=-0.021\pm0.0021$  and  $p_{66}=-0.068\pm0.003$ ). For the ADP crystals, the elastic stiffness coefficients are equal to  $C_{11}=62.0$ ,  $C_{12}=-5.0$ ,  $C_{13}=14.0$ ,  $C_{33}=30.0$ ,  $C_{44}=9.1$  and  $C_{66}=6.1$  GPa, while the elasto-optic coefficients

amount to  $p_{11}$  = 0.319,  $p_{12}$  = 0.277,  $p_{13}$  = 0.169,  $p_{31}$  = 0.197,  $p_{33}$  = 0.167,  $p_{44}$  = -0.058 and  $p_{66}$  = -0.091 [22]. All the optical data are taken for the same optical wavelength, 632.8 nm.

Anisotropy of the AW velocities in crystals can be analyzed using the Christoffel equation [25]:

$$C_{iikl}m_im_kp_l=\rho v^2p_i\,, (1)$$

where  $C_{ijkl}$  denote the components of elastic-stiffness tensor,  $m_j$  and  $m_k$  the components of the unit wave vector of AW, and  $p_i$  and  $p_l$  the components of the unit vector of displacement. The angle of deviation of the acoustic polarization from a purely longitudinal type is calculated basing on the Christoffel equation [25]:

$$\Delta = \zeta - \theta = \frac{1}{2} \arctan \frac{(C_{13} + C_{44}) \sin 2\theta}{(C_{11} - C_{44}) \cos^2 \theta + (C_{44} - C_{33}) \sin^2 \theta} - \theta , \qquad (2)$$

$$\Delta = \zeta - \theta = \frac{1}{2} \arctan \left( \frac{C_{12} + C_{66}}{C_{11} - C_{66}} \tan 2\theta \right) - \theta . \tag{3}$$

Here Eqs. (2) and (3) concern respectively the ac (or bc) and the ab planes,  $\theta$  implies the angle between the AW vector and the a (or b) axis, and  $\zeta$  the angle between the same axis and the displacement vector. The corresponding non-orthogonality of the quasi-transverse waves can, in principle, be calculated with the same formulae. The only difference is that an additive factor 90 deg should be added to the r.-h. s. of Eqs. (2) and (3). As seen from Fig. 1, the dependences of the angle of non-orthogonality on the wave vector orientation for the both crystals are similar for the ac plane and exactly the same for the ab plane. This angle reaches very high values (e.g.,  $\sim 37$  deg) in the ab plane.

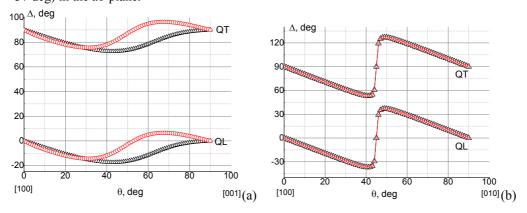


Fig. 1. Dependences of non-orthogonality angle on the AW vector orientation for KDP (circles) and ADP (triangles) crystals in the crystallographic planes *ac* (a) and *ab* (b).

The cross sections of the AW velocity surfaces are also similar for the KDP and ADP crystals (see Fig. 2 and Ref. [13] for the case of KDP). A schematic view of parts of the AW velocity surfaces, which are common for the both crystals, is presented in Fig. 3. There are five acoustic axes in the KDP and ADP crystals, which represent those propagation directions where the quasi-transverse AWs have the same phase velocity.

One of the acoustic axis is parallel to the crystallographic axis c, while the four other lie in the ab plane. The angular dependences of the AW velocities for the two quasi-transverse AWs for KDP and ADP touch each other in the principal ab plane. For the KDP crystals, the points where

these surfaces are touched lie at the angles  $V_1 = 18.35 \text{ deg} + n \times 90 \text{ deg}$  and  $V_2 = 71.65 \text{ deg} + n \times 90 \text{ deg}$  (n = 0, 1, 2, 3) with respect to the a axis (see Fig. 2). For the ADP crystals, the corresponding angles are  $V_1 = 13.90 \text{ deg} + n \times 90^\circ$  and  $V_2 = 76.10 \text{ deg} + n \times 90^\circ$ . In fact,

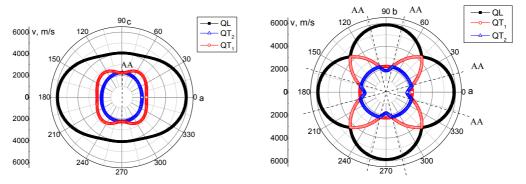


Fig. 2. Cross sections of AW velocity surfaces by the principal crystallographic planes for ADP crystals: AA denote the acoustic axes and QL the quasi-longitudinal waves.

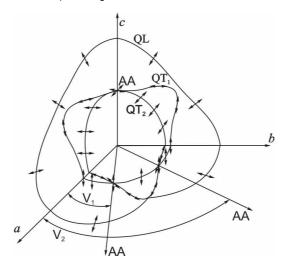


Fig. 3. Schematic view of parts of the AW velocity surfaces for ADP and KDP crystals.

the points mentioned above are nothing but polarization singularities, or topological defects of polarization orientation, with the strength equal to 1/2. This means that the AW polarization in the ab plane within the same AW velocity surface is switched by 90 deg when the wave vector crosses the acoustic axis direction. The acoustic axis parallel to the c axis also represents the polarization singularity. However, the strength of the topological defect of polarization distribution is equal to 1 in the latter case.

#### 2.2. Relations for the EEC

Using the procedures described in our recent work [20], one can derive the EEC for different types of AO interactions. In the mentioned work, the relations for the EEC have been obtained for the crystals of orthorhombic symmetry. These relations can be simplified for the tetragonal crystals that belong to the point symmetry groups 4/mmm, 4mm, 422 and  $\overline{4}2m$ , using the links among the elasto-optic coefficients  $p_{11}=p_{22}$ ,  $p_{12}=p_{21}$ ,  $p_{13}=p_{23}$ ,  $p_{31}=p_{32}$  and  $p_{44}=p_{55}$  imposed by the higher crystal symmetry.

The eigenvalues of the Christoffel equation can be found analytically only in case when the AWs propagate in the crystallographic planes. Thus, in the present work we consider the anisotropies of both the EEC and the AOFM only for the cases when the AWs propagate within the ac and ab planes. Notice that the EEC and AOFM anisotropies in the bc and ac planes are the same, due to symmetry conditions.

Let us consider the type I of isotropic AO interactions of ordinary optical wave with a quasi-longitudinal AW in the ac plane. With no accounting for polarization deviation from the purely longitudinal states, the EEC is written as

$$p_{ef}^{(1)} = p_{12}\cos^2\theta + p_{13}\sin^2\theta. \tag{4}$$

Taking such a deviation into account leads to a more complex relation,

$$p_{ef}^{(I)} = p_{12} \left( \cos^2 \theta \cos(\theta - \zeta) - \sin 2\theta \sin(\theta - \zeta) \right) + p_{13} \left( \sin^2 \theta \cos(\theta - \zeta) + \sin 2\theta \sin(\theta - \zeta) \right).$$
 (5)

It is seen that Eq. (5) is reduced to Eq. (4) at  $\zeta = \theta$ . For the *ab* plane the corresponding relations are given by

$$p_{ef}^{(I)} = \cos^{2}(\theta + \theta_{B}) \Big( p_{11} \cos^{2}\theta + p_{12} \sin^{2}\theta \Big) + \sin^{2}(\theta + \theta_{B}) \Big( p_{12} \cos^{2}\theta + p_{11} \sin^{2}\theta \Big),$$

$$-0.5 p_{66} \sin 2(\theta + \theta_{B}) \sin 2\theta$$

$$p_{ef}^{(I)} = \cos^{2}(\theta + \theta_{B}) \Big\{ \Big( p_{11} (\cos^{2}\theta \cos(\theta - \zeta) - \sin 2\theta \sin(\theta - \zeta) \Big) \Big\}$$

$$(6)$$

$$p_{ef}^{(I)} = \cos^{2}(\theta + \theta_{B}) \begin{cases} \left( p_{11}(\cos^{2}\theta\cos(\theta - \zeta) - \sin 2\theta\sin(\theta - \zeta)) \right) \\ + p_{12}\left(\sin^{2}\theta\cos(\theta - \zeta) + \sin 2\theta\sin(\theta - \zeta) \right) \end{cases}$$

$$+ \sin^{2}(\theta + \theta_{B}) \begin{cases} p_{12}\left(\cos^{2}\theta\cos(\theta - \zeta) - \sin 2\theta\sin(\theta - \zeta)\right) \\ + p_{11}\left(\sin^{2}\theta\cos(\theta - \zeta) + \sin 2\theta\sin(\theta - \zeta)\right) \end{cases}$$

$$(7)$$

$$-p_{66}\sin 2(\theta+\theta_B)(\cos 2\theta\sin(\theta-\zeta)+0.5\sin 2\theta\cos(\theta-\zeta))$$

If we deal with the type II of AO interactions of extraordinary optical wave with a quasi-longitudinal AW in the ac crystallographic plane, the EEC with no account for the polarization deviation from the purely longitudinal states can be presented as

$$p_{ef}^{(II)} = \cos^{2}(\theta + \theta_{B})(p_{11}\cos^{2}\theta + p_{13}\sin^{2}\theta) + \sin^{2}(\theta + \theta_{B})(p_{31}\cos^{2}\theta + p_{33}\sin^{2}\theta) + 0.5p_{44}\sin 2(\theta + \theta_{B})\sin 2\theta$$
(8)

The same relation written after considering properly for the deviation angle has the following form:

$$p_{ef}^{(II)} = \cos^{2}(\theta + \theta_{B}) \begin{cases} p_{11} \left( \cos^{2}\theta \cos(\theta - \zeta) - \sin 2\theta \sin(\theta - \zeta) \right) \\ + p_{13} \left( \sin^{2}\theta \cos(\theta - \zeta) + \sin 2\theta \sin(\theta - \zeta) \right) \end{cases}$$

$$+ \sin^{2}(\theta + \theta_{B}) \begin{cases} p_{31} \left( \cos^{2}\theta \cos(\theta - \zeta) - \sin 2\theta \sin(\theta - \zeta) \right) \\ + p_{33} \left( \sin^{2}\theta \cos(\theta - \zeta) + \sin 2\theta \sin(\theta - \zeta) \right) \end{cases}. \tag{9}$$

$$+p_{44}\sin 2(\theta+\theta_B)(\cos 2\theta\sin(\theta-\zeta)+0.5\sin 2\theta\cos(\theta-\zeta))$$

These relations for the ab plane read as

$$p_{ef}^{(II)} = p_{31}, (10)$$

$$p_{af}^{(II)} = p_{31} \cos(\theta - \zeta) . \tag{11}$$

Now let us consider AO interactions with the quasi-transverse AWs. The main peculiarity of these waves is the existence of acoustic axes along the c direction and in the ab plane. Notice that the transverse waves can propagate with arbitrary polarization orientations along the acoustic axes. The types III and IV of AO interactions with a so-called AW QT<sub>1</sub> can be realized in the interaction plane ac. The EEC for the type III of interactions of the ordinary optical wave is given by

$$p_{ef}^{(III)} = (p_{12} - p_{13})\sin 2\theta , \qquad (12)$$

if one neglects the angle of non-orthogonality. If this angle is accounted for, one arrives at

$$p_{ef}^{(III)} = p_{12} \left( \cos^2 \theta \cos(\zeta - \theta) - \sin 2\theta \sin(\zeta - \theta) \right) + p_{13} \left( \sin^2 \theta \cos(\zeta - \theta) + \sin 2\theta \sin(\zeta - \theta) \right).$$
(13)

It is evident that Eq. (13) reduces to Eq. (12) in the approximation of purely transverse AWs  $(\zeta = 270 + \theta)$ .

The type III of AO interactions cannot be fulfilled in the ab plane when the AW propagates at the angles  $0 \deg < \theta < V_1$  and  $V_2 < \theta < 90 \deg$  (i.e., in the first quadrant). The reason is that the appropriate elasto-optic coefficients are zero. However, the type III of AO interactions becomes possible when the AW propagates at the angles  $V_1 < \theta < V_2$  in the ab plane. Then the EEC is equal to

$$p_{ef}^{(III)} = p_{12} \left( \cos^2 \theta \cos(\zeta - \theta) - \sin 2\theta \sin(\zeta - \theta) \right) + p_{13} \left( \sin^2 \theta \cos(\zeta - \theta) + \sin 2\theta \sin(\zeta - \theta) \right)$$
(14)

with no considering of the non-orthogonality and

$$p_{ef}^{(III)} = \cos^{2}(\theta + \theta_{B}) \begin{cases} p_{11} \left(\cos^{2}\theta\cos(\zeta - \theta) - \sin 2\theta\sin(\zeta - \theta)\right) \\ + p_{12} \left(\sin^{2}\theta\cos(\zeta - \theta) + \sin 2\theta\sin(\zeta - \theta)\right) \end{cases} \\ + \sin^{2}(\theta + \theta_{B}) \begin{cases} p_{12} \left(\cos^{2}\theta\cos(\zeta - \theta) - \sin 2\theta\sin(\zeta - \theta)\right) \\ + p_{11} \left(\sin^{2}\theta\cos(\zeta - \theta) + \sin 2\theta\sin(\zeta - \theta)\right) \end{cases} \\ - p_{66} \sin 2(\theta + \theta_{B}) \left(0.5 \sin 2\theta\cos(\zeta - \theta) + \cos 2\theta\sin(\zeta - \theta)\right), \end{cases}$$
(15)

with accounting for this effect.

For the type IV of AO interactions of the extraordinary incident optical wave with the AW  $QT_1$ , we obtain the EEC for the ac plane of the form

$$p_{ef}^{(IV)} = \cos^{2}(\theta + \theta_{B})(p_{11} - p_{13})\sin 2\theta + \sin^{2}(\theta + \theta_{B})(p_{31} - p_{33})\sin 2\theta + p_{AA}\sin 2(\theta + \theta_{B})\cos 2\theta$$
(16)

with no account of the non-orthogonality. Proper consideration of the non-orthogonality effect yields in

$$p_{ef}^{(IV)} = \cos^{2}(\theta + \theta_{B}) \begin{cases} p_{11} \left( \cos^{2}\theta \cos(\zeta - \theta) - \sin 2\theta \sin(\zeta - \theta) \right) \\ + p_{13} \left( \sin^{2}\theta \cos(\zeta - \theta) + \sin 2\theta \sin(\zeta - \theta) \right) \end{cases}$$

$$+ \sin^{2}(\theta + \theta_{B}) \begin{cases} p_{31} \left( \cos^{2}\theta \cos(\zeta - \theta) - \sin 2\theta \sin(\zeta - \theta) \right) \\ + p_{33} \left( \sin^{2}\theta \cos(\zeta - \theta) + \sin 2\theta \sin(\zeta - \theta) \right) \end{cases}$$

$$+ p_{44} \sin 2(\theta + \theta_{B}) \left( 0.5 \sin 2\theta \cos(\zeta - \theta) + \cos 2\theta \sin(\zeta - \theta) \right)$$

$$(17)$$

The type IV of AO interactions in the ab interaction plane is impossible for the AW QT<sub>1</sub> propagating at the angles  $0 \deg < \theta < V_1$  and  $V_2 < \theta < 90 \deg$  (i.e., in the first quadrant), since the appropriate elasto-optic coefficients are zero. This is no longer true when the AWs propagate in the ab plane at the angles  $V_1 < \theta < V_2$ . Then the type IV of AO interactions can be fulfilled, and the EEC becomes as follows (note that the condition  $p_{31} = p_{32}$  holds true):

$$p_{ef}^{(IV)} = (p_{31} - p_{32})\sin 2\theta = 0, (18)$$

$$p_{ef}^{(IV)} = p_{31} \cos(\zeta - \theta)$$
, (19)

Eqs. (18) and (19) refer respectively to the cases when the non-orthogonality effect is taken into account and neglected.

In the ac crystallographic plane, the types V and VI of AO interactions with a so-called AW QT<sub>2</sub> cannot be implemented for the crystals that belong to point group  $\overline{4}2m$ , since the appropriate elasto-optic coefficients are equal to zero. On the other hand, the type V of AO interactions with the ordinary optical wave becomes possible in the ab plane when the AW propagates at the angles in the first quadrant (0 deg  $<\theta< V_1$  and  $V_2<\theta<90$  deg ). Then the EEC reads as

$$p_{ef}^{(V)} = (p_{11} - p_{12})\sin 2\theta \cos 2(\theta + \theta_B) + p_{66}\sin 2(\theta + \theta_B)\cos 2\theta , \qquad (20)$$

and

$$p_{ef}^{(V)} = \cos^{2}(\theta + \theta_{B}) \begin{cases} p_{11} \left(\cos^{2}\theta \cos(\zeta - \theta) - \sin 2\theta \sin(\zeta - \theta)\right) \\ + p_{12} \left(\sin^{2}\theta \cos(\zeta - \theta) + \sin 2\theta \sin(\zeta - \theta)\right) \end{cases}$$

$$+ \sin^{2}(\theta + \theta_{B}) \begin{cases} p_{12} \left(\cos^{2}\theta \cos(\zeta - \theta) - \sin 2\theta \sin(\zeta - \theta)\right) \\ + p_{11} \left(\sin^{2}\theta \cos(\zeta - \theta) + \sin 2\theta \sin(\zeta - \theta)\right) \end{cases}$$

$$- p_{66} \sin 2(\theta + \theta_{B}) \left(0.5 \sin 2\theta \cos(\zeta - \theta) + \cos 2\theta \sin(\zeta - \theta)\right),$$

$$(21)$$

when the non-orthogonality effect is respectively neglected by and accounted for. The type V of AO interactions is impossible when the AW propagates in the ab plane at the angles  $V_1 < \theta < V_2$ .

When the AW propagates at the angles  $0 \deg < \theta < V_1$  and  $V_2 < \theta < 90 \deg$  in the ab plane, we obtain the EEC (  $p_{31} = p_{32}$  )

$$p_{ef}^{(VI)} = (p_{31} - p_{32})\sin 2\theta = 0$$
 (22)

and

$$p_{ef}^{(VI)} = p_{31}\cos(\zeta - \theta)$$
, (23)

which are valid for the cases of accounting for and neglecting by the non-orthogonality. The type VI of AO interactions cannot be realized when the AW propagates in the ab plane at the angles  $V_1 < \theta < V_2$ .

Let us now consider the anisotropic AO diffraction. In the principal crystallographic planes, this diffraction can be implemented only with purely transverse AWs. The AO interactions with the purely transverse AW QT<sub>2</sub> become possible in the ac plane, and the interactions with the AWs QT<sub>1</sub> or QT<sub>2</sub> can be implemented in the ab plane. In the latter case, we should deal with the propagation angles  $0 \deg < \theta < V_1$  and  $V_2 < \theta < 90 \deg$  for the AW QT<sub>1</sub> or the angles  $V_1 < \theta < V_2$  for the AW QT<sub>2</sub>. The anisotropic AO interactions in the crystallographic planes cannot be realized with any other AWs, since the corresponding elasto-optic coefficients are zero. The EEC for the

type VIII of AO interactions with the AW QT<sub>1</sub> in the ab plane can be written as

$$p_{ef}^{(VIII)} = p_{44}\cos(\theta - \alpha), \qquad (24)$$

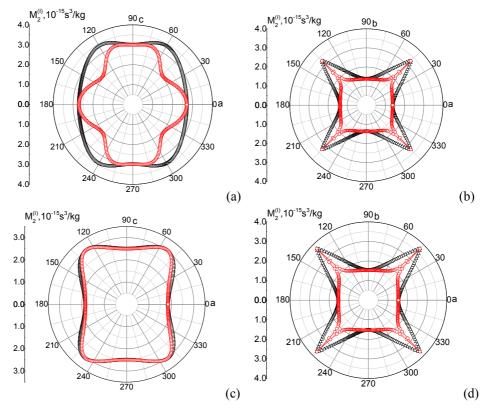
where the AW propagates at the angles  $0 \deg < \theta < V_1$  and  $V_2 < \theta < 90 \deg$ , and  $\alpha$  denotes the angle between the a axis and the wave vector of the incident optical wave. For the type IX of AO interactions of the AW QT<sub>2</sub> in the ac plane, the EEC is given by

$$p_{ef}^{(IX)} = p_{66}\cos\theta\sin\alpha + p_{44}\sin\theta\cos\alpha. \tag{25}$$

In case of the AW propagation angles  $V_1 < \theta < V_2$ , the type IX of AO interactions with the AW QT<sub>2</sub> in the ab plane is again described by Eq. (24).

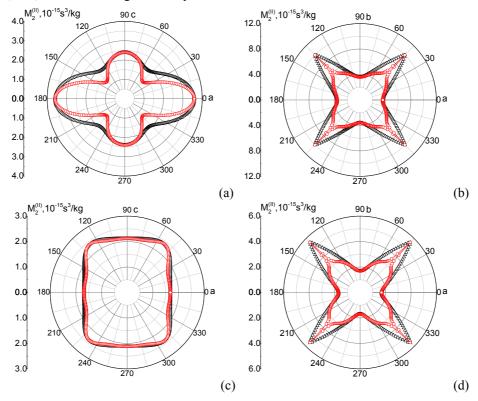
#### 3. Results and discussion

The AOFM anisotropy for the type I of AO interactions in the ADP and KDP crystals is illustrated in Fig. 4. Consideration of the non-orthogonality effect in the crystallographic plane ab changes notably the angular behaviour of the AOFM for the both crystals (cf. panels (b) and (d) in Fig. 4). However, for the ac plane the non-orthogonality of AWs influences the AOFM anisotropy only for the ADP crystals (Fig. 4a). On the contrary, the AOFMs for the KDP crystals found under conditions of accounting for or neglecting by the AW non-orthogonality behave almost identically with changing  $\theta$  angle (see Fig. 4c). This fact can be explained when examining Eqs. (4) and (5) and the elasto-optic coefficients  $p_{12}$  and  $p_{13}$  for the both crystals. In particular, the parameters  $p_{12}$  and  $p_{13}$  are almost the same for KDP and differ notably for ADP.



**Fig. 4.** Dependences of AOFM on the angle  $\theta$  for the type *I* of AO interactions in the *ac* (a, c) and *ab* (b, d) planes for ADP (a, b) and KDP (c, d) crystals: open triangles and circles correspond respectively to neglecting and taking the AW non-orthogonality into account.

The maximal AOFM value for the ADP crystals peculiar for the type I of AO interactions is equal to  $3.3 \times 10^{-15} \, \text{s}^3/\text{kg}$ . It is reached at the angles  $\theta = 45$ , 135, 225 and 315 deg in the ab plane. Since this maximum is very sharp, it would be more convenient from the practical viewpoint to deal with the interaction geometry in the ac plane ( $\theta = 90$  or 180 deg). The AOFM under these conditions is only a little smaller,  $3.0 \times 10^{-15} \, \text{s}^3/\text{kg}$ . The maximal AOFM for the KDP crystals  $(3.7 \times 10^{-15} \, \text{s}^3/\text{kg})$  can be achieved in the ab plane at  $\theta = 45$ , 135, 225 and 315 deg. Only slightly lower AOFM value,  $3.0 \times 10^{-15} \, \text{s}^3/\text{kg}$ , can be reached at somewhat better practical conditions  $\theta = 53$ , 127, 233 and 307 deg in the ac plane.

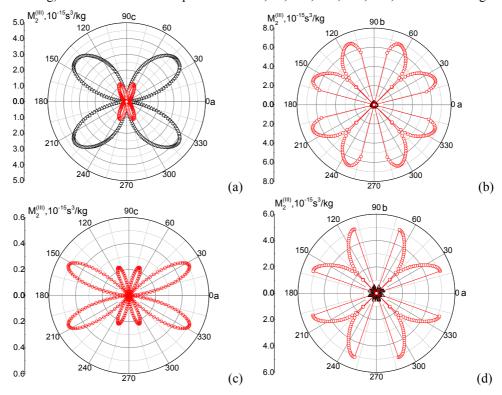


**Fig. 5.** Dependences of AOFM on the angle  $\theta$  for the type II of AO interactions in the ac (a, c) and ab (b, d) planes for ADP (a, b) and KDP (c, d) crystals: open triangles and circles correspond respectively to neglecting and accounting for AW non-orthogonality.

The AOFM anisotropy at the type II of AO interactions shown in Fig. 5 is similar to that found for the type I. Namely, the AOFM anisotropies for ADP in the ac plane differs noticeably under the conditions of accounting for and neglecting by the AW non-orthogonality, although they are almost the same for the KDP (cf. Fig. 5a with Fig. 5c). Such behaviour for the KDP crystals follows from close values of the elasto-optic coefficients  $p_{11}$ ,  $p_{13}$ ,  $p_{31}$  and  $p_{33}$  included into Eqs. (8) and (9). The difference among these coefficients is more pronounced for the ADP crystals. The anisotropy of AOFM in the ab plane is caused mainly by the anisotropy of AW slowness (see Fig. 5b, d). The maximal AOFM for the ADP crystals is equal to  $10.0 \times 10^{-15}$  s<sup>3</sup>/kg, being achieved in the ab plane at  $\theta = 45$ , 135, 225 and 315 deg. The maximal AOFM value for the KDP crystals,  $5.6 \times 10^{-15}$  s<sup>3</sup>/kg, is reached under the same conditions.

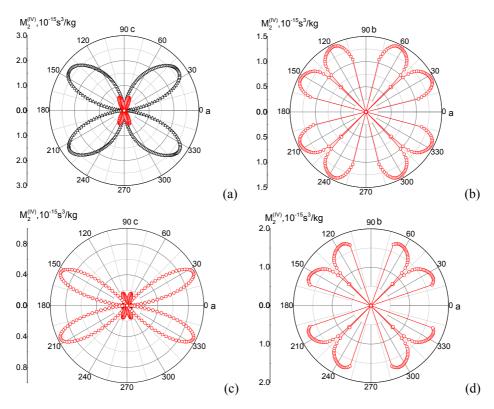
In case of the type III of AO interactions in the ac plane, the AOFM calculated with consideration of the AW non-orthogonality is higher than that derived when disregarding it (see

Fig. 6a). However, the AOFM of KDP in the ac plane calculated when the non-orthogonality effect is neglected is negligibly small, when compared with that calculated with taking the effect into account (see Fig. 6c). Let us remind that the elasto-optic tensor components  $p_{12} = 0.249 \pm 0.013$ and  $p_{13} = 0.242 \pm 0.012$  for KDP are almost the same, especially if one takes their inevitable experimental errors into account. Then Eq. (12) can be rewritten as  $p_{ef}^{(III)} = (p_{12} - p_{13})\sin 2\theta \approx 0$ . This is the reason why the AOFM for KDP in the ac plane is almost equal to zero when the AW non-orthogonality is not considered. It follows from Eq. (13) that, under the condition  $p_{12} \approx p_{13}$ , the EEC can be rewritten as  $p_{ef}^{(III)} \approx p_{12} \cos(\zeta - \theta)$ . Hence, accounting for the angle of nonorthogonality in the KDP crystals leads to nonzero AOFM value in the crystallographic plane ac. The mentioned elasto-optic coefficient for the ADP crystals differs almost as twice, whereas the angle of non-orthogonality is somewhat higher than that for the KDP crystals. This results in decreasing AOFM in the ac plane for ADP, when the deviation of AW polarization from the purely transverse types is considered. Neglecting by the angle of non-orthogonality in the ab plane for the both crystals leads to a critical decrease in the AOFM (see Fig. 6b, d), since the elasto-optic coefficients  $p_{11}$  and  $p_{12}$  for the ADP and KDP crystals are close to each other, while the coefficient  $p_{66}$  is very small. As a result, the  $p_{ef}^{(III)}$  parameter given by Eq. (14) is small enough. It follows from Eq. (15) that accounting for the angle of non-orthogonality imposes increasing AOFM for the both crystals in the ab plane. The maximal AOFM value for ADP  $(6.9 \times 10^{-15} \text{ s}^3/\text{kg})$  is reached in the ab plane at  $\theta = 23$ , 67, 113, 157, 203, 247, 293 and 337 deg. The maximal AOFM for KDP  $(5.1 \times 10^{-15} \text{ s}^3/\text{kg})$  is reached in the same plane at  $\theta = 20, 70, 110, 160, 200, 250, 290$  and 340 deg.



**Fig. 6.** Dependences of AOFM on the angle  $\theta$  for the type *III* of AO interactions in the *ac* (a, c) and *ab* (b, d) planes for ADP (a, b) and KDP (c, d) crystals: open triangles and circles correspond respectively to neglecting by and accounting for AW non-orthogonality.

For the type *IV* of AO interactions, the behaviour of AOFM (see Fig. 7) is similar to that calculated for the type *III*. However, the EEC for the *ab* plane is equal to zero (see Eq. (18)) when the non-orthogonality is neglected (see Fig. 7b, d). On the other hand, consideration of this effect leads to deviation of the EEC from zero, according to Eq. (19). The maximal AOFM value for the ADP crystals,  $1.4 \times 10^{-15}$  s<sup>3</sup>/kg, is achieved in the *ab* plane at  $\theta = 27$ , 63, 117, 153, 207, 243, 297 and 333 deg. For KDP crystals, we have the maximal AOFM value  $1.8 \times 10^{-15}$  s<sup>3</sup>/kg in the same interaction plane at  $\theta = 25$ , 65, 115, 155, 205, 245, 295 and 335 deg.



**Fig. 7.** Dependences of AOFM on the angle  $\theta$  for the type *IV* of AO interactions in the *ac* (a, c) and *ab* (b, d) planes for ADP (a, b) and KDP (c, d) crystals: open triangles and circles correspond respectively to neglecting and accounting for AW non-orthogonality.

The types V and VI of AO interactions can be realized only in the crystallographic plane ab. Then the angular behaviours of the AOFM for the both crystals are similar (Fig. 8). Almost a zero AOFM value obtained when the non-orthogonality is neglected is caused by close elasto-optic coefficients that define the EEC (see Eqs. (20)–(23)). The maximal AOFM values at the type V of AO interactions are equal to  $5.2 \times 10^{-15} \, \text{s}^3/\text{kg}$  and  $5.1 \times 10^{-15} \, \text{s}^3/\text{kg}$ . They are reached in the ab plane at  $\theta = 13$ , 77, 103, 166, 193, 257, 283 and 346 deg for ADP and at  $\theta = 18$ , 71, 108, 162, 198, 252, 288 and 341 deg for KDP. Concerning the type VI of AO interactions, we obtain the maximal AOFM values equal to  $1.0 \times 10^{-15} \, \text{s}^3/\text{kg}$  for ADP (at  $\theta = 13$ , 77, 103, 166, 193, 257, 283 and 346 deg) and  $1.6 \times 10^{-15} \, \text{s}^3/\text{kg}$  for KDP (at  $\theta = 18$ , 71, 108, 162, 198, 252, 288 and 341 deg).

Let us consider the anisotropic AO diffraction in the ADP and KDP crystals. Fig. 9 shows the dependences of AOFM on the angle  $\alpha + \gamma$ . We remind that  $\alpha$  is the angle of incidence of the

light wave (i.e., the angle between the wave vector of the incident optical wave and the a axis) and  $\gamma$  the diffraction angle (i.e., the angle between the wave vectors of the incident and diffracted optical waves). The type *VIII* of AO interactions can be realized in the crystallographic plane ab. The maximal AOFM value for the ADP crystals,  $2.1 \times 10^{-15} \, \text{s}^3/\text{kg}$  (see Fig. 9a), which is peculiar for the type *VIII* of AO interactions, is reached at  $\alpha = 90 \, \text{deg}$ ,  $\alpha + \gamma = 82 \, \text{or} \, 98 \, \text{deg}$ , and  $\gamma = +8 \, \text{or} \, -8 \, \text{deg}$ . The maximal AOFM for the KDP crystals (see Fig. 9b) is reached under the same conditions, although its value is somewhat lower,  $1.8 \times 10^{-15} \, \text{s}^3/\text{kg}$ .

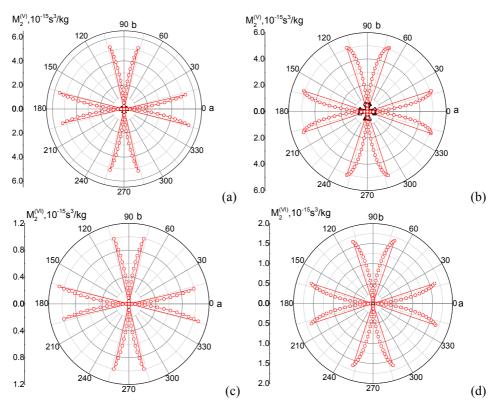


Fig. 8. Anisotropy of AOFM for the types V(a, b) and VI(c, d) of AO interactions for ADP (a, c) and KDP (b, d) crystals.

Using the type *IX* of AO interactions (see Fig. 9c, d), we obtain the maximal AOFM  $9.5 \times 10^{-15} \, \text{s}^3/\text{kg}$  for the ADP crystals. It is reached at almost collinear AO interactions, i.e. at  $\alpha = 90 \, \text{deg}$  and  $\alpha + \gamma = 89 \, \text{or} \, 91 \, \text{deg}$ , and  $\gamma = +1 \, \text{or} \, -1 \, \text{deg}$ . The maximal AOFM for the KDP crystals,  $5.3 \times 10^{-15} \, \text{s}^3/\text{kg}$ , is achieved under the same conditions.

Let us examine the collinear diffraction as a particular case of anisotropic diffraction. The collinear diffraction can be implemented at  $\gamma=0$  or 90 deg. For the type *VIII* of AO interactions (see Fig. 10a), the maximal AOFM for the ADP crystals is equal to  $0.5 \times 10^{-15} \, \text{s}^3/\text{kg}$ . This value is peculiar for the *ab* plane at  $\alpha=68$ , 112, 248 and 292 deg. The maximal AOFM for the KDP crystals (see Fig. 10b) is the same and occurs under identical conditions. Finally, the maximal AOFM for the ADP crystals and the type *IX* of AO interactions in the *ac* plane is equal to  $4.4 \times 10^{-15} \, \text{s}^3/\text{kg}$  at  $\alpha=41$ , 139, 221 and 319 deg. It is equal to  $1.2 \times 10^{-15} \, \text{s}^3/\text{kg}$  for KDP and occurs at  $\alpha=38$ , 142, 218 and 322 deg.

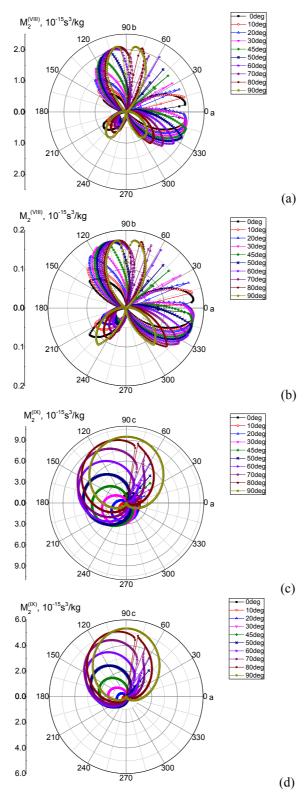


Fig. 9. Dependences of AOFM for the types  $\emph{VIII}$  (a, b) and  $\emph{IX}$  (c, d) of AO interactions in ADP (a, c) and KDP (b, d) crystals on the angle  $\alpha+\gamma$ , as calculated at different incidence angles  $\alpha$  of optical wave.

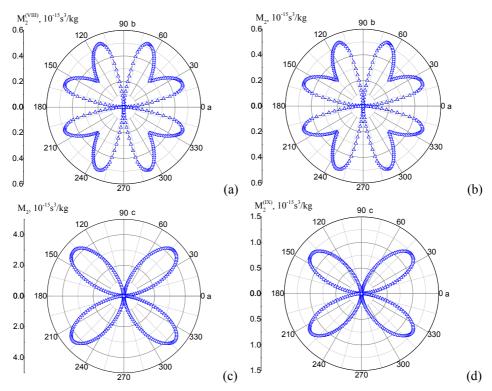


Fig. 10. Dependences of AOFM on the angle  $\alpha$  at the collinear diffraction at the types VIII (a, b) and IX (c, d) of AO interactions in ADP (a, c) and KDP (b, d) crystals.

Table 1. Maximal AOFM values calculated for the ADP and KDP crystals at different types of isotropic AO interactions.

Type of AO interactions	Ι		II		III		IV		V		VI	
					K	DP						
Angle $\theta$ ,	45,	135,	45,	135,	20,	70,	25,	65,	18,	71,	18,	71,
deg, and	225	and	225	and	110,	160,	115,	155,	108,	162,	108,	162,
interaction	315,		315,		200,	250,	205,	245,	198,	252,	198,	252,
plane	ab pl	ane	ab pl	ane	290	and	295	and	288	and	288	and
_					340,		335,		341,		341,	
					ab pla	ane	ab pla	ane	ab pla	ane	ab pla	ane
AOFM, 10 <sup>-15</sup> s <sup>3</sup> /kg	3	.7	5	.6	5	.1	1	.8	5	.1	1	.6
					A.	DP						
Angle $\theta$ ,	45,	135,	45,	135,	23,	67,	27,	63,	13,	77,	13,	77,
deg, and	225	and	225	and	113,	157,	117,	153,	103,	166,	103,	166,
interaction	315,		315,		203,	247,	207,	243,	193,	257,	193,	257,
plane	ab pl	ane	ab pl	ane	293	and	297	and	283	and	283	and
					337,		333,		346,		346,	
					<i>ab</i> plane		<i>ab</i> plane		ab plane		ab plane	
AOFM, $10^{-15}  \text{s}^3/\text{kg}$	3.3		10.0		6.9		1.4		5.2		1.0	

As seen from Table 1, the principal AOFM maximum for KDP at the isotropic diffraction amounts to  $5.6 \times 10^{-15} \, \text{s}^3/\text{kg}$ . This value is achieved at the type II of AO interactions with the quasi-longitudinal AW in the crystallographic plane ab. The principal AOFM maximum for ADP,  $10.0 \times 10^{-15} \, \text{s}^3/\text{kg}$ , is reached for the same type of AO interactions and in the same interaction plane. Notice that this value is the highest AOFM found for all the types of AO interactions in the crystallographic planes for the both crystals.

Table 2. Maximal AOFM values for the ADP and KDP crystals at different types of anisotropic AO interactions.

Type of AO	VIII	IX	VIII	IX
interactions			(collinear)	(collinear)
		KDP		
Angles $\alpha$ , deg;	90; +8 or -8,	90; +1  or  -1,	68, 112, 248 and	38, 142, 218 and
$\gamma$ , deg, and	ab plane	ac plane	292; 0,	322; 0,
interaction plane			ab plane	ac plane
AOFM,	1.8	5.3	0.5	1.2
$10^{-15}  \mathrm{s}^3/\mathrm{kg}$				
		ADP		
Angles $\alpha$ , deg;	90; +8 or –8,	90; +1  or  -1,	68, 112, 248 and	41, 139, 221 and
$\gamma$ , deg, and	<i>ab</i> plane	ac plane	292; 0,	319; 0,
interaction plane			<i>ab</i> plane	ac plane
AOFM,	2.1	9.5	0.5	4.4
$10^{-15}  \mathrm{s}^3/\mathrm{kg}$				

In case of the anisotropic diffraction (see Table 2), the highest AOFM value can be reached at the type IX of AO interactions with the AW QT<sub>2</sub> in the ac plane. The highest AOFMs are equal to  $5.3 \times 10^{-15} \, \text{s}^3/\text{kg}$  and  $9.5 \times 10^{-15} \, \text{s}^3/\text{kg}$  for KDP and ADP, respectively. The latter value is comparable with the highest one achieved at the isotropic diffraction. The collinear diffraction is also characterized by the highest AOFMs at the type IX of AO interactions. These values are equal to  $1.2 \times 10^{-15} \, \text{s}^3/\text{kg}$  and  $4.4 \times 10^{-15} \, \text{s}^3/\text{kg}$  for KDP and ADP, respectively.

## 4. Conclusions

In the present work we have derived the analytical relations for the EEC with accounting for the angle of non-orthogonality of AW polarization in case of the crystals belonging to the symmetry group  $\overline{4}2m$ . The same relations are valid for the other point groups of tetragonal system, such as 4/mmm, 422 and 4mm. The above relations can also be used for the point symmetry groups 6/mmm, 6mm, 622 and  $\overline{6}m2$  of hexagonal system, as well as for the Curie symmetry groups  $\infty 2$ ,  $\infty mm$  and  $\infty / m$ , with taking into account the additional relations  $p_{66} = (p_{11} - p_{12})/2$  and  $C_{66} = (C_{11} - C_{12})/2$  valid for the latter groups. We have analyzed the anisotropy of AOFM for the KDP and ADP crystals within their crystallographic planes in the cases when the effect of non-orthogonality of AW polarizations is neglected by or accounted for. We have found that consideration of the effect mentioned above changes significantly the AOFM value. This concerns almost all the types of AO interactions, except for the types I and II of interactions with the quasi-longitudinal AWs in KDP. Moreover, in some cases the non-orthogonality of AW polarizations can lead to appearance of AO interactions, although this interaction is forbidden at the zero angle of non-orthogonality. The latter fact concerns the types IV and VI of interactions with the AWs QT<sub>1</sub> and QT<sub>2</sub>, respectively.

The geometries of AO diffraction at which the maximal AOFM values have to be observed are obtained for all of the nine types of AO interactions, including the particular case of collinear diffraction. We have demonstrated that the principal AOFM maximum for the KDP crystals is equal to  $5.6 \times 10^{-15} \, \text{s}^3/\text{kg}$  at the isotropic diffraction. The corresponding value for the ADP crystals amounts to  $10.0 \times 10^{-15} \, \text{s}^3/\text{kg}$ . Note that the latter value represents the highest AOFM calculated for all the types of AO interactions in the crystallographic planes for the both crystals. It can be achieved at the type II of AO interactions with the quasi-longitudinal AW in the crystallographic plane ab. The highest AOFM value for the case of anisotropic AO diffraction can be reached at the type IX of interactions with the AW QT<sub>2</sub> in the ac plane. It is equal to  $5.3 \times 10^{-15} \, \text{s}^3/\text{kg}$  and  $9.5 \times 10^{-15} \, \text{s}^3/\text{kg}$  for KDP and ADP, respectively. The latter value is comparable with the highest AOFM achieved at the isotropic diffraction. The collinear diffraction is also characterized by the highest AOFMs at the type IX of AO interactions. These values are equal respectively to  $1.2 \times 10^{-15} \, \text{s}^3/\text{kg}$  and  $4.4 \times 10^{-15} \, \text{s}^3/\text{kg}$  for KDP and ADP. Finally, the ADP crystals reveal higher AOFMs at any of the isotropic, anisotropic or collinear AO diffractions, if compared with the KDP crystals.

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Анотація. У роботі одержано аналітичні вирази для ефективних пружнооптичних коефіцієнтів з урахуванням неортогональності поляризації акустичних хвиль (АХ) у кристалах, що належать до груп симетрії 4/тт, 422 і 4тт. Проаналізовано анізотропію коефіцієнта акустооптичної (AO) якості для кристалів KDP і ADP в кристалографічних площинах. Цю анізотропію одержано за умов, коли ефект неортогональності АХ знехтуваний або врахований. Показано, що належне врахування кута неортогональності АХ істотно змінює величину коефіцієнта акустооптичної якості. Встановлено геометрії АОдифракції, для яких можна одержати максимальні значення коефіцієнта акустооптичної якості для всіх дев'яти типів АО-взаємодій, включаючи ті, що стосуються колінеарної дифракції. Продемонстровано, що найбільший максимум якості дорівню $\epsilon$   $10.0 \times 10^{-15} \, c^3/\kappa z$  і має місце для кристалів ADP. Його можна досягти для AO-взаємодій типу II із квазіпоздовжніми АХ у кристалографічній площині ав. Найвище значення коефіцієнта акустоптичної якості, характерне для анізотропної АО-дифракції в АDP, складає  $9.5 \times 10^{-15} \, c^3/\kappa$ г і досягається для AO-взаємодій типу IX із так званою  $AX \, OT_2$  у площині ас. Нарешті, найвищий коефіцієнт якості для випадку колінеарної дифракції дорівнює  $4,4\times10^{-15}~c^3/\kappa$ г і також властивий кристалам ADP.