Anisotropy of acousto-optic figure of merit in tetragonal crystals with accounting for non-orthogonality of acoustic eigenwave polarizations.
1. The cases of KH₂PO₄ and NH₄H₂PO₄ crystals

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Abstract. In the present work we derive analytical relations for the effective elasto-optic 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 KH₂PO₄ and NH₄H₂PO₄ 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 non-orthogonality 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 NH₄H₂PO₄ is equal to 10.0×10^{-15} s³/kg. 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 peculiar for the anisotropic AO diffraction in NH₄H₂PO₄, 9.5×10^{-15} s³/kg, can be reached at the type IX of AO interactions with a so-called AW QT₂ in the *ac* plane. Finally, the highest AOFM found for the case of collinear diffraction, 4.4×10^{-15} s³/kg, is also inherent to the NH₄H₂PO₄ crystals. KH₂PO₄ is characterized by somewhat lower AOFM values.

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 TeO₂ and the point symmetry group 422 [1]), of which AO figure of merit (AOFM) is as high as 1200×10^{-15}s³/kg [2]. We remind that the AOFM is determined by the relationship $M_2 = n^6 p_{ef}^2 / \rho v^3$, where ρ 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, Li₂B₄O₇ 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 GW/cm² (according to our measurements [3]) or even 40 GW/cm² (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 3.44×10^{-15} s^3/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.

KH_2PO_4 (KDP) and $NH_4H_2PO_4$ (ADP) crystals characterized by the point symmetry group $\overline{4}2m$ 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 7.1×10^{-15} s^3/kg [13]. According to the literature data, the AOFM for the ADP crystals amounts to 6.43×10^{-15} s^3/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$_2$, Li$_2$B$_4$O$_7$ and NaBi(MoO$_4$)$_2$. 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$_2$, Li$_2$B$_4$O$_7$ and NaBi(MoO$_4$)$_2$, 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/m^3 and $\rho = 1803$ kg/m^3, respectively [1]. In our calculations, we use the elastic-stiffness coefficients of the KDP crystals obtained in Ref. [23] ($C_{11} = 71.4 \pm 0.8$, $C_{12} = -4.9 \pm 1.0$, $C_{13} = 12.9 \pm 0.3$, $C_{33} = 56.15 \pm 0.3$, $C_{44} = 12.7 \pm 0.1$ and $C_{66} = 6.24 \pm 0.05$ GPa) and the elasto-optic coefficients determined in Ref. [24] ($p_{11} = 0.238 \pm 0.024$, $p_{12} = 0.249 \pm 0.013$, $p_{13} = 0.242 \pm 0.012$, $p_{31} = 0.227 \pm 0.011$, $p_{33} = 0.242 \pm 0.024$, $p_{44} = -0.021 \pm 0.0021$ and $p_{66} = -0.068 \pm 0.003$). 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]:

$$C_{ijkl}m_j m_k p_l = \rho v^2 p_i, \tag{1}$$

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]:

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

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

Here Eqs. (2) and (3) concern respectively the *ac* (or *bc*) and the *ab* planes, θ implies the angle between the AW vector and the *a* (or *b*) axis, and ζ 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., ~ 37 deg) in the *ab* plane.
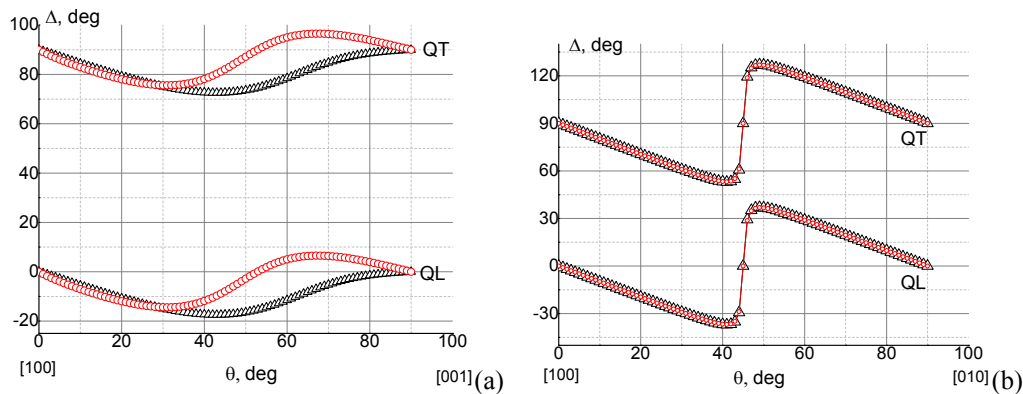

Fig. 1. Dependences of non-orthogonality angle on the AW vector orientation for KDP (circles) and ADP (triangles) crystals in the crystallographic planes *ac* (a) and *ab* (b).
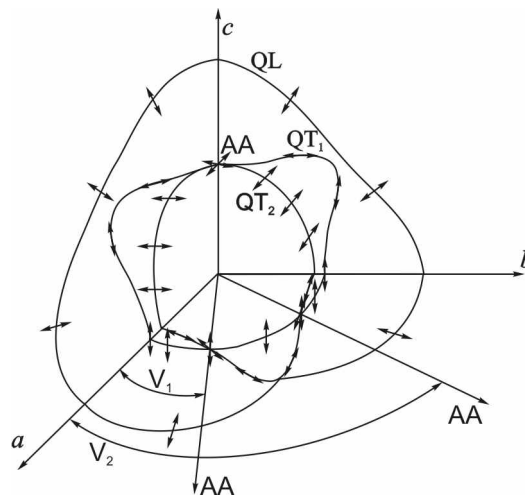
The cross sections of the AW velocity surfaces are also similar for the KDP and ADP crystals (see Fig. 2 and Ref. [13] for the case of KDP). A schematic view of parts of the AW velocity surfaces, which are common for the both crystals, is presented in Fig. 3. There are five acoustic axes in the KDP and 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 $V_1 = 18.35$ deg $+ n \times 90$ deg and $V_2 = 71.65$ deg $+ n \times 90$ deg ($n = 0, 1, 2, 3$) with respect to the a axis (see Fig. 2). For the ADP crystals, the corresponding angles are $V_1 = 13.90$ deg $+ n \times 90°$ and $V_2 = 76.10$ deg $+ n \times 90°$. In fact,
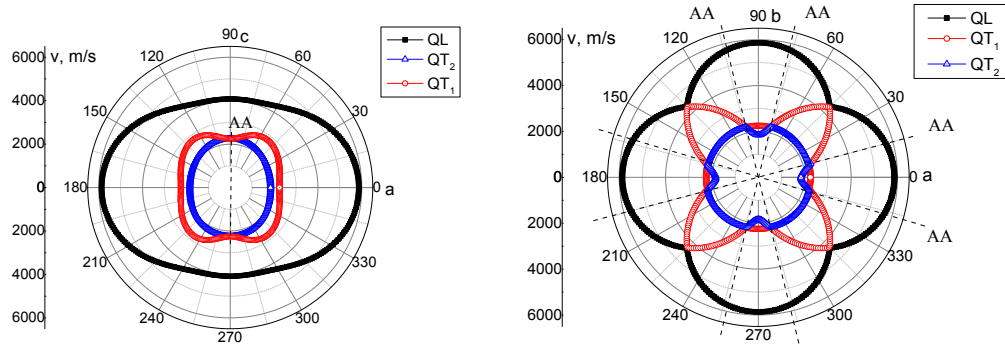
Fig. 2. Cross sections of AW velocity surfaces by the principal crystallographic planes for ADP crystals: AA denote the acoustic axes and QL the quasi-longitudinal waves.

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

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

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

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

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

$$p_{ef}^{(I)} = \cos^2(\theta + \theta_B) \left(p_{11} \cos^2 \theta + p_{12} \sin^2 \theta \right) + \sin^2(\theta + \theta_B) \left(p_{12} \cos^2 \theta + p_{11} \sin^2 \theta \right) ,$$
$$-0.5 p_{66} \sin 2(\theta + \theta_B) \sin 2\theta \tag{6}$$

$$p_{ef}^{(I)} = \cos^2(\theta + \theta_B) \left\{ \begin{matrix} \left(p_{11}(\cos^2 \theta \cos(\theta - \zeta) - \sin 2\theta \sin(\theta - \zeta)) \right) \\ + p_{12} \left(\sin^2 \theta \cos(\theta - \zeta) + \sin 2\theta \sin(\theta - \zeta) \right) \end{matrix} \right\}$$
$$+ \sin^2(\theta + \theta_B) \left\{ \begin{matrix} p_{12} \left(\cos^2 \theta \cos(\theta - \zeta) - \sin 2\theta \sin(\theta - \zeta) \right) \\ + p_{11} \left(\sin^2 \theta \cos(\theta - \zeta) + \sin 2\theta \sin(\theta - \zeta) \right) \end{matrix} \right\} . \tag{7}$$
$$- p_{66} \sin 2(\theta + \theta_B) \left(\cos 2\theta \sin(\theta - \zeta) + 0.5 \sin 2\theta \cos(\theta - \zeta) \right)$$

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

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

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

$$p_{ef}^{(II)} = \cos^2(\theta + \theta_B) \left\{ \begin{matrix} p_{11} \left(\cos^2 \theta \cos(\theta - \zeta) - \sin 2\theta \sin(\theta - \zeta) \right) \\ + p_{13} \left(\sin^2 \theta \cos(\theta - \zeta) + \sin 2\theta \sin(\theta - \zeta) \right) \end{matrix} \right\}$$
$$+ \sin^2(\theta + \theta_B) \left\{ \begin{matrix} p_{31} \left(\cos^2 \theta \cos(\theta - \zeta) - \sin 2\theta \sin(\theta - \zeta) \right) \\ + p_{33} \left(\sin^2 \theta \cos(\theta - \zeta) + \sin 2\theta \sin(\theta - \zeta) \right) \end{matrix} \right\} . \tag{9}$$
$$+ p_{44} \sin 2(\theta + \theta_B) \left(\cos 2\theta \sin(\theta - \zeta) + 0.5 \sin 2\theta \cos(\theta - \zeta) \right)$$

These relations for the *ab* plane read as

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

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

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$_1$ can be realized in the interaction plane ac. The EEC for the type III of interactions of the ordinary optical wave is given by

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

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

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

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

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

with no considering of the non-orthogonality and

$$\begin{aligned} p_{ef}^{(III)} = \cos^2(\theta+\theta_B)\left\{ \begin{array}{l} p_{11}\left(\cos^2\theta\cos(\zeta-\theta) - \sin 2\theta\sin(\zeta-\theta)\right) \\ + p_{12}\left(\sin^2\theta\cos(\zeta-\theta) + \sin 2\theta\sin(\zeta-\theta)\right) \end{array} \right\} \\ + \sin^2(\theta+\theta_B)\left\{ \begin{array}{l} p_{12}\left(\cos^2\theta\cos(\zeta-\theta) - \sin 2\theta\sin(\zeta-\theta)\right) \\ + p_{11}\left(\sin^2\theta\cos(\zeta-\theta) + \sin 2\theta\sin(\zeta-\theta)\right) \end{array} \right\} \\ - p_{66}\sin 2(\theta+\theta_B)\left(0.5\sin 2\theta\cos(\zeta-\theta) + \cos 2\theta\sin(\zeta-\theta)\right), \end{aligned} \tag{15}$$

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

$$\begin{aligned} p_{ef}^{(IV)} = \cos^2(\theta+\theta_B)(p_{11} - p_{13})\sin 2\theta + \sin^2(\theta+\theta_B)(p_{31} - p_{33})\sin 2\theta \\ + p_{44}\sin 2(\theta+\theta_B)\cos 2\theta \end{aligned} \tag{16}$$

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

$$\begin{aligned} p_{ef}^{(IV)} = \cos^2(\theta+\theta_B)\left\{ \begin{array}{l} p_{11}\left(\cos^2\theta\cos(\zeta-\theta) - \sin 2\theta\sin(\zeta-\theta)\right) \\ + p_{13}\left(\sin^2\theta\cos(\zeta-\theta) + \sin 2\theta\sin(\zeta-\theta)\right) \end{array} \right\} \\ + \sin^2(\theta+\theta_B)\left\{ \begin{array}{l} p_{31}\left(\cos^2\theta\cos(\zeta-\theta) - \sin 2\theta\sin(\zeta-\theta)\right) \\ + p_{33}\left(\sin^2\theta\cos(\zeta-\theta) + \sin 2\theta\sin(\zeta-\theta)\right) \end{array} \right\} \\ + p_{44}\sin 2(\theta+\theta_B)\left(0.5\sin 2\theta\cos(\zeta-\theta) + \cos 2\theta\sin(\zeta-\theta)\right) \end{aligned} \tag{17}$$

The type *IV* of AO interactions in the *ab* interaction plane is impossible for the AW QT$_1$ 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):

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

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

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$_2$ 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

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

and

$$\begin{aligned}
p_{ef}^{(V)} = {}& \cos^2(\theta + \theta_B)\left\{\begin{array}{l} p_{11}\left(\cos^2\theta\cos(\zeta - \theta) - \sin 2\theta\sin(\zeta - \theta)\right) \\ + p_{12}\left(\sin^2\theta\cos(\zeta - \theta) + \sin 2\theta\sin(\zeta - \theta)\right)\end{array}\right\} \\
& + \sin^2(\theta + \theta_B)\left\{\begin{array}{l} p_{12}\left(\cos^2\theta\cos(\zeta - \theta) - \sin 2\theta\sin(\zeta - \theta)\right) \\ + p_{11}\left(\sin^2\theta\cos(\zeta - \theta) + \sin 2\theta\sin(\zeta - \theta)\right)\end{array}\right\} \\
& - p_{66}\sin 2(\theta + \theta_B)\left(0.5\sin 2\theta\cos(\zeta - \theta) + \cos 2\theta\sin(\zeta - \theta)\right),
\end{aligned} \tag{21}$$

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 ($p_{31} = p_{32}$)

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

and

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

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$_2$ become possible in the *ac* plane, and the interactions with the AWs QT$_1$ or QT$_2$ 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$_1$ or the angles $V_1 < \theta < V_2$ for the AW QT$_2$. 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_1 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 α 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_2 in the *ac* plane, the EEC is given by

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

In case of the AW propagation angles $V_1 < \theta < V_2$, the type *IX* of AO interactions with the AW QT_2 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 θ 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.
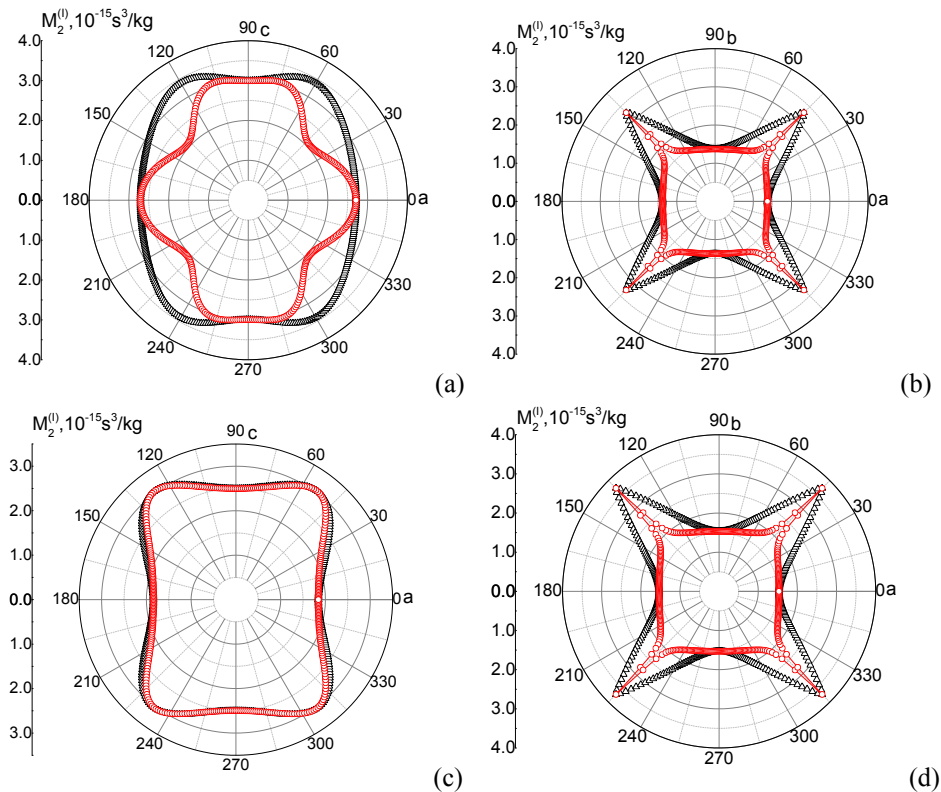
Fig. 4. Dependences of AOFM on the angle θ for the type *I* of AO interactions in the *ac* (a, c) and *ab* (b, d) planes for ADP (a, b) and KDP (c, d) crystals: open triangles and circles correspond respectively to neglecting and taking the AW non-orthogonality into account.

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.
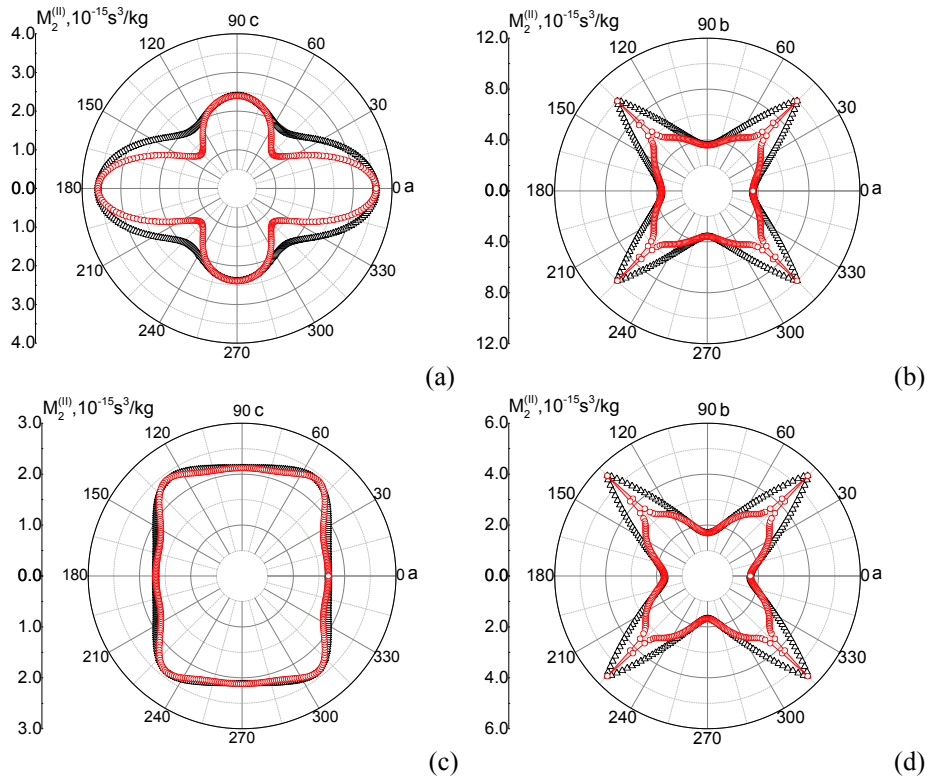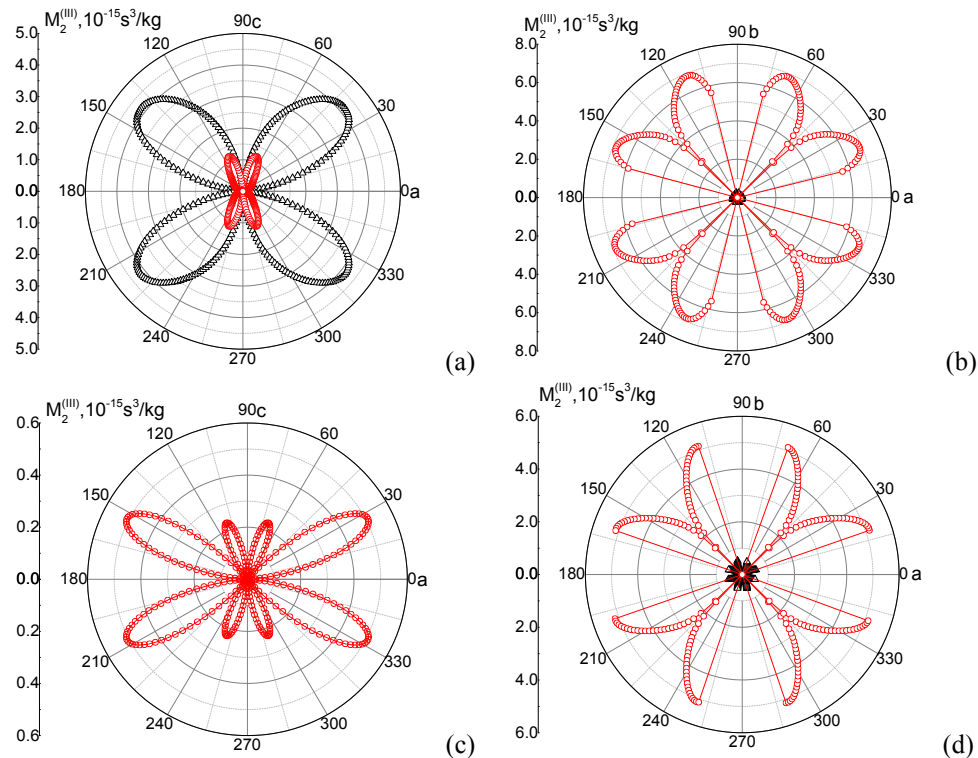

Fig. 5. Dependences of AOFM on the angle θ for the type *II* of AO interactions in the *ac* (a, c) and *ab* (b, d) planes for ADP (a, b) and KDP (c, d) crystals: open triangles and circles correspond respectively to neglecting and accounting for AW non-orthogonality.

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 $10.0 \times 10^{-15}\,\text{s}^3/\text{kg}$, being achieved in the *ab* plane at $\theta = 45, 135, 225$ and 315 deg. The maximal AOFM value for the KDP crystals, $5.6 \times 10^{-15}\,\text{s}^3/\text{kg}$, is reached under the same conditions.
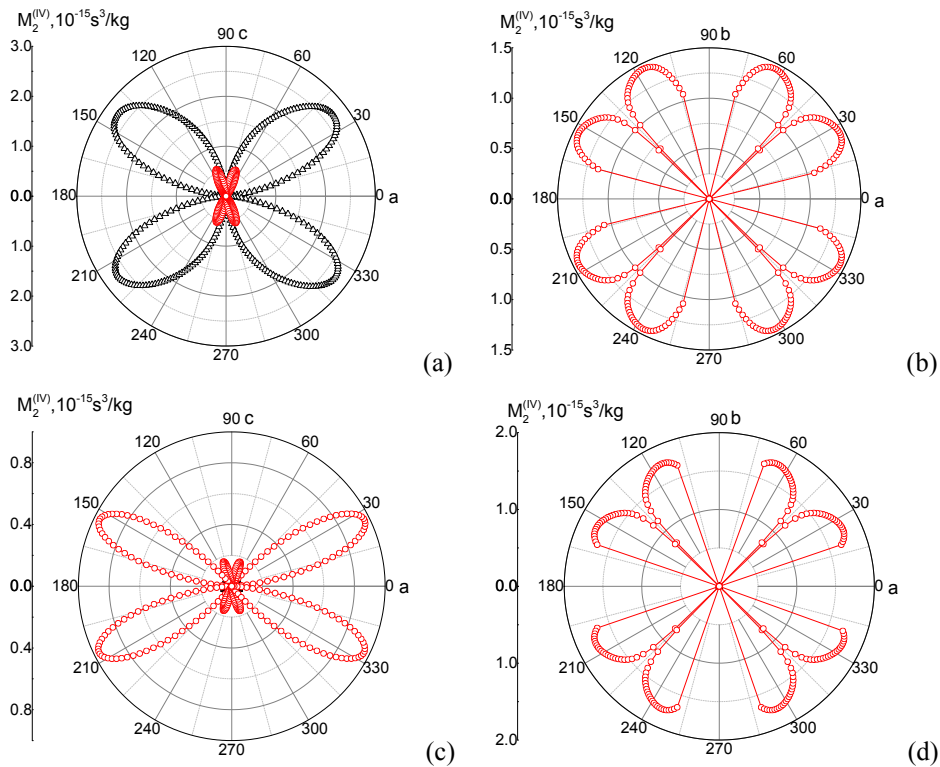
In case of the type *III* of AO interactions in the *ac* plane, the AOFM calculated with consideration of the AW non-orthogonality is higher than that derived when disregarding it

(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 non-orthogonality 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 (6.9×10^{-15} s^3/kg) is reached in the *ab* plane at $\theta = 23, 67, 113, 157, 203, 247, 293$ and 337 deg. The maximal AOFM for KDP (5.1×10^{-15} s^3/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 θ 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, 1.4×10^{-15} s^3/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 1.8×10^{-15} s^3/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 θ 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 5.2×10^{-15} s^3/kg and 5.1×10^{-15} s^3/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 1.0×10^{-15} s^3/kg for ADP (at $\theta = 13, 77, 103, 166, 193, 257, 283$ and 346 deg) and 1.6×10^{-15} s^3/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 α is the angle of incidence of the

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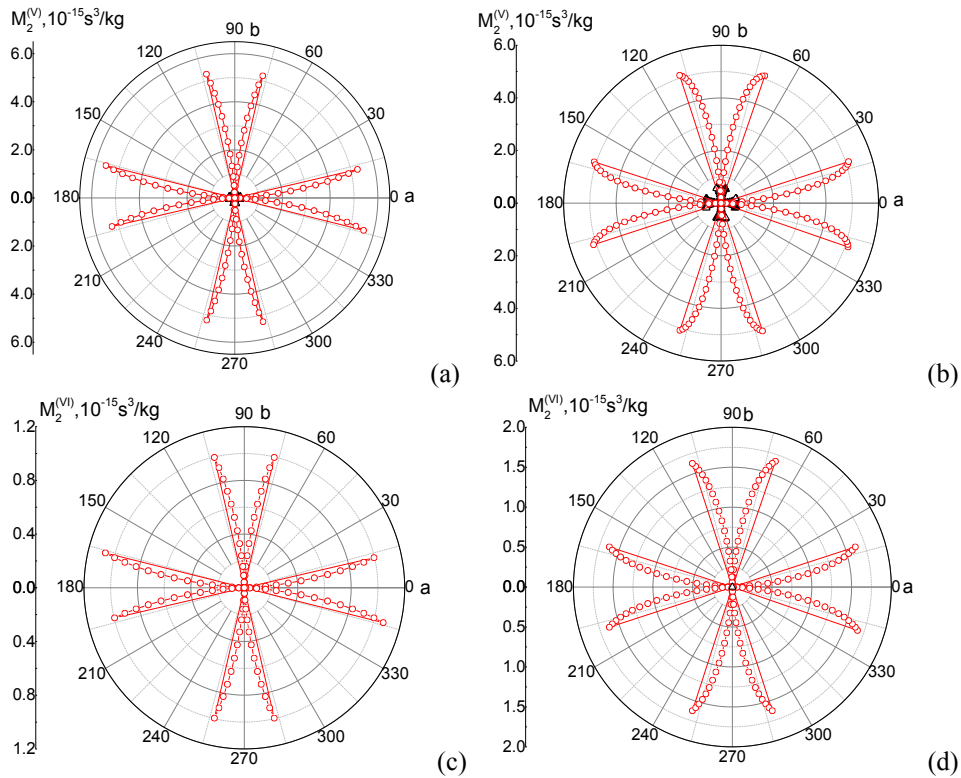

Fig. 8. Anisotropy of AOFM for the types *V* (a, b) and *VI* (c, d) of AO interactions for ADP (a, c) and KDP (b, d) crystals.

Using the type *IX* of AO interactions (see Fig. 9c, d), we obtain the maximal AOFM 9.5×10^{-15} s^3/kg for the ADP crystals. It is reached at almost collinear AO interactions, i.e. at $\alpha = 90$ deg and $\alpha + \gamma = 89$ or 91 deg, and $\gamma = +1$ or -1 deg. The maximal AOFM for the KDP crystals, 5.3×10^{-15} s^3/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 0.5×10^{-15} s^3/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 4.4×10^{-15} s^3/kg at $\alpha = 41, 139, 221$ and 319 deg. It is equal to 1.2×10^{-15} s^3/kg for KDP and occurs at $\alpha = 38, 142, 218$ and 322 deg.
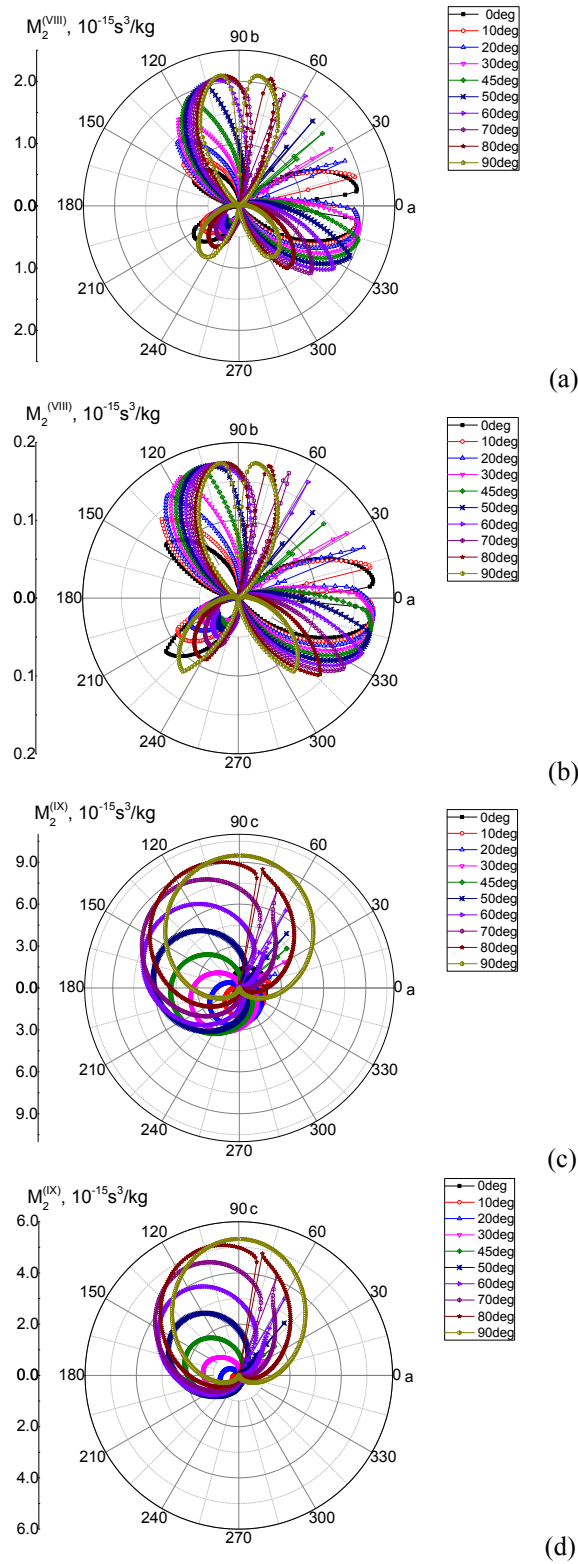
Fig. 9. Dependences of AOFM for the types *VIII* (a, b) and *IX* (c, d) of AO interactions in ADP (a, c) and KDP (b, d) crystals on the angle $\alpha + \gamma$, as calculated at different incidence angles α of optical wave.
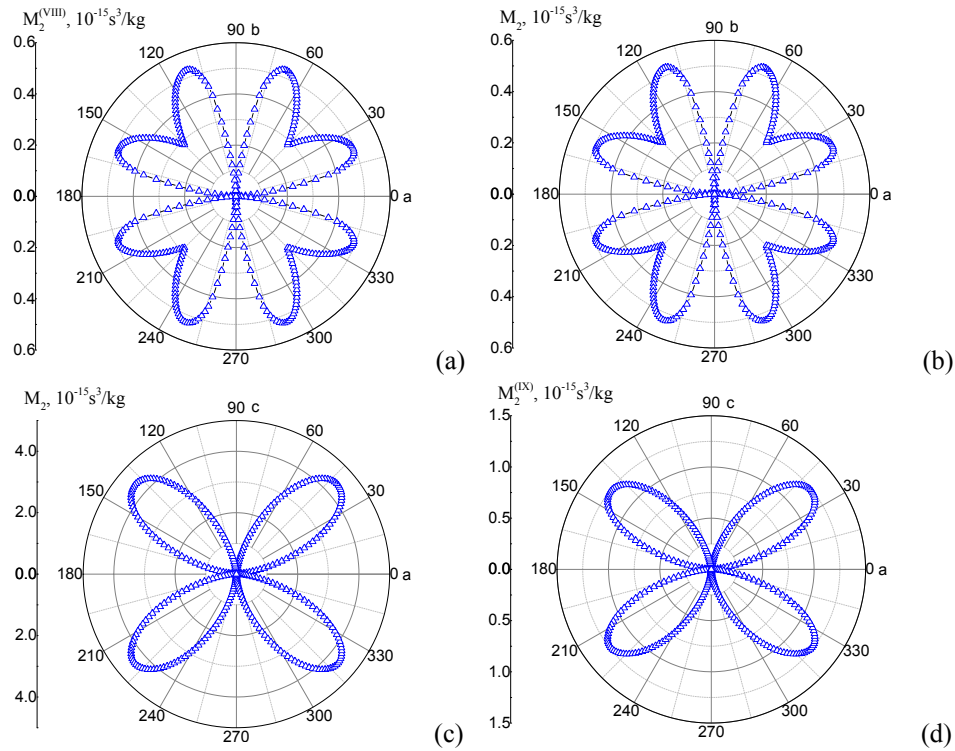
Fig. 10. Dependences of AOFM on the angle α at the collinear diffraction at the types *VIII* (a, b) and *IX* (c, d) of AO interactions in ADP (a, c) and KDP (b, d) crystals.

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

Type of AO interactions	*I*	*II*	*III*	*IV*	*V*	*VI*
	KDP					
Angle θ, deg, and interaction plane	45, 135, 225 and 315, *ab* plane	45, 135, 225 and 315, *ab* plane	20, 70, 110, 160, 200, 250, 290 and 340, *ab* plane	25, 65, 115, 155, 205, 245, 295 and 335, *ab* plane	18, 71, 108, 162, 198, 252, 288 and 341, *ab* plane	18, 71, 108, 162, 198, 252, 288 and 341, *ab* plane
AOFM, 10^{-15} s^3/kg	3.7	5.6	5.1	1.8	5.1	1.6
	ADP					
Angle θ, deg, and interaction plane	45, 135, 225 and 315, *ab* plane	45, 135, 225 and 315, *ab* plane	23, 67, 113, 157, 203, 247, 293 and 337, *ab* plane	27, 63, 117, 153, 207, 243, 297 and 333, *ab* plane	13, 77, 103, 166, 193, 257, 283 and 346, *ab* plane	13, 77, 103, 166, 193, 257, 283 and 346, *ab* plane
AOFM, 10^{-15} s^3/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 5.6×10^{-15} s^3/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, 10.0×10^{-15} s^3/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 interactions	$VIII$	IX	$VIII$ (collinear)	IX (collinear)
KDP				
Angles α, deg; γ, deg, and interaction plane	90; +8 or −8, ab plane	90; +1 or −1, ac plane	68, 112, 248 and 292; 0, ab plane	38, 142, 218 and 322; 0, ac plane
AOFM, 10^{-15} s^3/kg	1.8	5.3	0.5	1.2
ADP				
Angles α, deg; γ, deg, and interaction plane	90; +8 or −8, ab plane	90; +1 or −1, ac plane	68, 112, 248 and 292; 0, ab plane	41, 139, 221 and 319; 0, ac plane
AOFM, 10^{-15} s^3/kg	2.1	9.5	0.5	4.4

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$_2$ in the ac plane. The highest AOFMs are equal to 5.3×10^{-15} s^3/kg and 9.5×10^{-15} s^3/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 1.2×10^{-15} s^3/kg and 4.4×10^{-15} s^3/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 ∞2, ∞mm and ∞/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$_1$ and QT$_2$, 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 5.6×10^{-15} s^3/kg at the isotropic diffraction. The corresponding value for the ADP crystals amounts to 10.0×10^{-15} s^3/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$_2$ in the *ac* plane. It is equal to 5.3×10^{-15} s^3/kg and 9.5×10^{-15} s^3/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 1.2×10^{-15} s^3/kg and 4.4×10^{-15} s^3/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/mmm, 422 і 4mm. Проаналізовано анізотропію коефіцієнта акустооптичної (АО) 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 АО-дифракції, 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 дорівнює $10,0 \times 10^{-15}$ с³/кг і має місце для кристалів ADP. Його можна досягти для АО-взаємодій типу II із квазі-поздовжніми АХ у кристалографічній площині ab. Найвище значення коефіцієнта акустоптичної якості, характерне для анізотропної АО-дифракції в ADP, складає $9,5 \times 10^{-15}$ с³/кг і досягається для АО-взаємодій типу IX із так званою АХ QT_2 у площині ac. Нарешті, найвищий коефіцієнт якості для випадку колінеарної дифракції дорівнює $4,4 \times 10^{-15}$ с³/кг і також властивий кристалам ADP.