

## EFFECT OF MAGNETIC FIELD ON THE ACOUSTO-OPTIC DIFFRACTION EFFICIENCY IN $\text{NaBi}(\text{MoO}_4)_2$ CRYSTALS

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Received: 09.07.2025

**Abstract.** The anisotropic acousto-optic (AO) diffraction in  $\text{NaBi}(\text{MoO}_4)_2$  crystals under the effect of magnetic field is considered in the present work. It has been shown that the induced circular birefringence by the magnetic field due to the Faraday effect can lead to the enhancement of efficiency of AO diffraction if the incident optical wave is circularly polarized and propagates along optical axis. At the interaction with the quasi-longitudinal acoustic wave, the AO figure of merit changes from a value of  $0.3 \times 10^{-15} \text{ s}^3/\text{kg}$  at zero magnetic field to  $8 \times 10^{-15} \text{ s}^3/\text{kg}$  when the magnetic field is applied. The actual experimental geometry for further validation of our theoretical predictions is proposed.

**Keywords:** acousto-optic figure of merit, Faraday effect, magnetic field, double sodium bismuth molybdate crystal

**UDC:** 535.4, 534.2

**DOI:** 10.3116/16091833/Ukr.J.Phys.Opt.2025.03057

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### 1. Introduction

Double sodium bismuth molybdate crystals  $[\text{NaBi}(\text{MoO}_4)_2]$  belong to the point group of symmetry  $4/m$  and possess the scheelite mineral structure [1-3]. The unit cell parameters of  $\text{NaBi}(\text{MoO}_4)_2$  crystals under normal conditions are equal to  $a = b = 0.5274 \text{ nm}$  and  $c = 1.1578 \text{ nm}$ , and their unit cell contains two formula units [3]. The  $\text{NaBi}(\text{MoO}_4)_2$  crystals are optically negative with the refractive indices equal to  $n_o = 2.295$  and  $n_e = 2.198$  at the wavelength of  $633 \text{ nm}$  [4]. These crystals are interesting from the viewpoints of their acousto-optic (AO) properties [5]. According to the results of work [3], the  $\text{NaBi}(\text{MoO}_4)_2$  crystals undergo ferroelastic phase transition with the change of spatial symmetry  $I4_1/a \rightarrow FI2_1/a$  at  $T_c = 241 \text{ K}$ . This phase transition is accompanied by the appearance of shear strains of their unit cell in the crystallographic plane  $ab$  of the tetragonal lattice set and by small displacements of  $\text{Na}^+$  and  $\text{Bi}^{3+}$  cations from their high-symmetry positions [3]. As a result, the phase transition in  $\text{NaBi}(\text{MoO}_4)_2$  crystals seems to be of a displacement type. If the acoustic properties of double sodium molybdate crystals [6] are considered, the minimum of acoustic wave (AW) velocity for the quasi-transverse AW  $\text{QT}_1$  ( $1948 \text{ m/s}$ ) propagated under the angle  $62^\circ$  to the  $a$  axis in the  $ab$  plane and polarized in the same plane is observed. Most probably, this AW is conjugated with the soft acoustic mode, which is responsible for this phase transition. On the other hand, it is known that the ferroelastic crystals are the best AO materials [7]. Moreover, usually the most efficient geometry of AO interaction is the geometry at which optical waves interact with the slowest AW. Therefore, in the present work, we will analyze the AO interaction with the  $\text{QT}_1$  AW in the  $a'c$  plane, where the  $a'$  axis is rotated on the angle  $62^\circ$  with respect to  $a$  axis in the  $ab$  crystallographic plane.

Additionally, we will analyze the AO diffraction at the interaction with quasi-longitudinal AW (QL) and QT<sub>2</sub> AW.

As it has been shown in our recent works [8-10], the efficiency of AO diffraction can be enhanced in the case when the ellipticity of polarization of the incident optical wave matches the polarization of the eigen optical wave. The elliptical polarization of the incident wave involves the additional elasto-optic (EO) tensor components in the effective EO coefficient. The additional term, which contains these tensor components, is multiplied by the ellipticity of the incident optical wave. As a result, the effective EO coefficient and therefore the AO figure of merit increase with increasing ellipticity. The highest ellipticity value, i.e.,  $\pm 1$ , is peculiar for the circularly polarized optical wave. This is why anisotropic AO interaction with circularly polarized optical waves increases the AO figure of merit from  $(600-800) \times 10^{-15} \text{ s}^3/\text{kg}$  for the linearly polarized optical waves to  $1200 \times 10^{-15} \text{ s}^3/\text{kg}$  for the circularly polarized optical waves in TeO<sub>2</sub> crystal [11]. Notice that this phenomenon has been experimentally proved by us using the ellipticity of incident wave modulation [12].

The eigen optical waves are circularly polarized in optically active media when linear birefringence equals zero. However, the circularly polarized optical waves can occur not only in the media with natural optical activity, but also when external fields induce optical activity. In the present case, we will consider optical activity induced by the magnetic field due to the Faraday effect. It should be noted that we have considered the same effect in recent works on the example of other crystals [13,14]. However, here the NaBi(MoO<sub>4</sub>)<sub>2</sub> crystals are chosen as the material for our subsequent experimental verification of the analysis made in the present work. Therefore, we will analyze the anisotropic AO Bragg diffraction in NaBi(MoO<sub>4</sub>)<sub>2</sub> crystals under the condition of magnetic field-induced circular birefringence.

## 2. Method of analysis

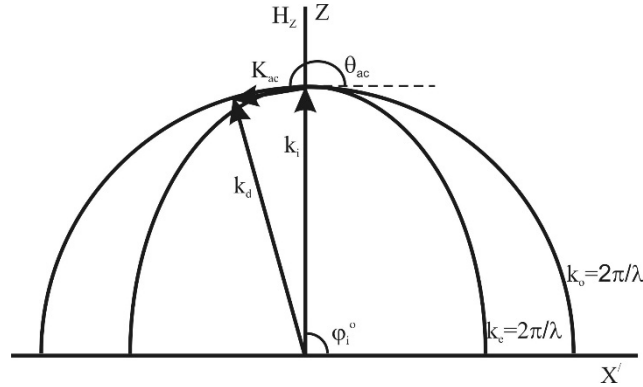
Let the incident optical wave propagate along the Z crystal physical axis, which is parallel to the crystallographic axis *c*. Since we will consider the anisotropic AO diffraction, the dependencies of the effective EO coefficient and AO figure of merit will be built on the diffraction angle  $\gamma$ , which is coupled with the direction of propagation of the AW by the relation:

$$\theta_{ac}(\varphi_i + \gamma) = a \tan \left\{ \frac{n_o \sin(\varphi_i) - \frac{n_o n_e \sin(\varphi_i + \gamma)}{\sqrt{n_o^2 \cos^2(\varphi_i + \gamma) + n_e^2 \sin^2(\varphi_i + \gamma)}}}{n_o \cos(\varphi_i) - \frac{n_o n_e \cos(\varphi_i + \gamma)}{\sqrt{n_o^2 \cos^2(\varphi_i + \gamma) + n_e^2 \sin^2(\varphi_i + \gamma)}}} \right\} \bigg|_{\varphi_i = \pi/2}, \quad (1)$$

$$= a \tan \left\{ \frac{\sqrt{n_o^2 \sin^2 \gamma + n_e^2 \cos^2 \gamma} - n_e \cos \gamma}{n_e \sin \gamma} \right\}$$

where  $\varphi_i$  is the angle between *X'* axis and the direction of propagation of the incident optical wave (*X'* is parallel to the *a'* axis),  $\theta_{ac}$  is the angle between the direction of propagation of AW and the *X'* axis. In the above relation, we neglect the change in refractive indices caused by the magnetic field due to the Faraday effect (i.e. circular birefringence), as this contribution is quite small. Let's start our analysis with the AO interaction with quasi-transverse QT<sub>1</sub> AW, which, at propagation in the *ab* plane, is polarized within the same plane.

As mentioned above, at propagation in the  $ab$  plane, the AW acquires a minimal velocity when the acoustic wavevector is parallel to the  $X'$  axis. Therefore, we will consider AO diffraction in the  $X'Z$  plane (Fig. 1).



**Fig. 1.** Schematic view of the phase matching conditions at the anisotropic AO interaction with QT1 AW.

The deformation tensor components caused by this AW in  $X'Y'Z$  coordinate system can be written as:

$$e'_4 = \sin\theta_{ac} \text{ and } e'_6 = \cos\theta_{ac}. \quad (2)$$

We have neglected the angle of non-orthogonality here because it is less than 1 deg. The effective EO coefficient derived by the standard procedure (see e.g. [8]) in  $X'Y'Z$  coordinate system is as follows:

$$p'_{eff} = 0.5 \left[ (p'_{16}(\varphi_Z) + p'_{66}(\varphi_Z)) \cos^2 \theta_{ac} + p'_{45} \sin^2 \theta_{ac} \right] + 0.5 \chi^2 \left[ (p'_{66}(\varphi_Z) - p'_{16}(\varphi_Z)) \cos^2 \theta_{ac} + p'_{44} \sin^2 \theta_{ac} \right], \quad (3)$$

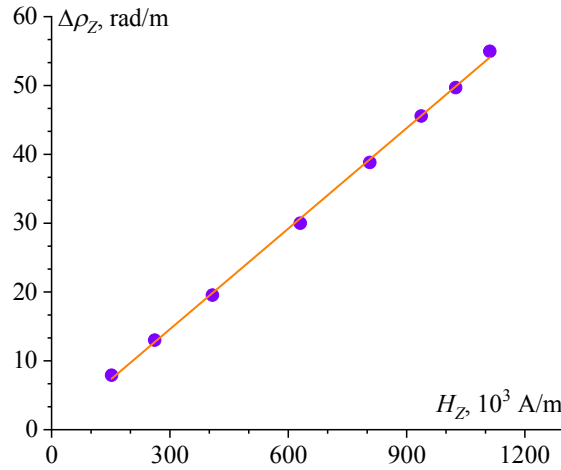
where

$$p'_{16}(\varphi_Z) = (p_{16} - p_{61}) \cos^2 2\varphi_Z - 0.5(p_{11} - 0.5p_{12} - p_{66}) \sin 4\varphi_Z - p_{61}, \quad (4)$$

$$p'_{66}(\varphi_Z) = p_{66} - 0.5(p_{16} + p_{61}) \sin^2 4\varphi_Z + 0.5(p_{11} - p_{12} - 2p_{66}) \sin^2 2\varphi_Z, \quad (5)$$

and  $\varphi_Z$  is the angle between  $X$  and  $X'$  axes. The EO coefficients in the  $XYZ$  coordinate system has been taken from Ref. [15] and are equal to:  $p_{11}=0.196 \pm 0.005$ ,  $p_{12}=0.159 \pm 0.007$ ,  $p_{13}=0.189 \pm 0.003$ ,  $p_{31}=0.154 \pm 0.002$ ,  $p_{33}=0.228 \pm 0.003$ ,  $p_{44}=0.019 \pm 0.002$ ,  $p_{66}=-0.044 \pm 0.004$ ,  $p_{45}=0.017 \pm 0.004$ ,  $p_{16}=-0.023 \pm 0.004$  and  $p_{61}=0.035 \pm 0.012$ . At  $\varphi_Z=62^\circ$   $p'_{16}=0.071$  and  $p'_{66}=-0.0005$ . The ellipticity of the incident optical wave  $|\chi|=1$  at it propagation along optical axis at the presence of magnetic field and  $\chi=0$  at switching off magnetic field. The dependence of specific rotatory power on the magnetic field strength for  $\text{NaBi}(\text{MoO}_4)_2$  crystals, obtained experimentally at the light wavelength  $\lambda=632.8$  nm is presented in Fig. 2.

Calculated Verdet constant is equal to  $V_F = (\Delta\rho_Z / H_Z) / \mu_0 = (38.73 \pm 0.20) \text{ rad}/(\text{T} \times \text{m})$ , where  $\mu_0 = 4\pi \times 10^{-7} \text{ H/m}$  is the vacuum magnetic permeability. Then the Faraday coefficient is equal to  $F_{33} = (\lambda \Delta\rho_Z / H_Z) / \pi n_o^3 = (7.974 \pm 0.041) \times 10^{-13} \text{ m/A}$ .



**Fig. 2.** Dependence of specific rotatory power on the magnetic field strength  $H_Z$  for  $\text{NaBi}(\text{MoO}_4)_2$  crystals ( $\lambda = 632.8 \text{ nm}$ ). Violet circles correspond to an experimental data, orange straight line correspond to an approximation by a linear function  $\Delta\rho_Z = KH_Z$ .

The  $\text{QT}_1$  AW velocity is calculated with using of the formula:

$$v_{\text{QT}_1}(\theta_{ac}) = \sqrt{\frac{C'_{66}(\varphi_Z)\cos^2\theta_{ac} + C_{44}\sin^2\theta_{ac}}{\rho}}, \quad (6)$$

where

$$C'_{66}(\varphi_Z) = C_{66} - C_{16}\sin 4\varphi_Z + 0.5(C_{11} - C_{12} - 2C_{66})\sin^2 2\varphi_Z, \quad (7)$$

and  $C_{11}=105.40\pm 0.33$ ,  $C_{12}=47.96\pm 15.84$ ,  $C_{13}=35.58\pm 1.41$ ,  $C_{16}=-10.64\pm 1.07$ ,  $C_{33}=90.22\pm 0.41$ ,  $C_{44}=25.40\pm 0.07$  and  $C_{66}=37.51\pm 0.33 \text{ GPa}$  [6] and  $\rho = 5690 \text{ kg/m}^3$  [3].

Let us consider the AO interaction with QL AW that propagates in the plane  $X'Z$  rotated by the angle of 17 deg relative to the  $X$  axis. At the propagation in the  $XY$  plane, this AW acquires minimal velocity, i.e., 4156 m/s. The relation for the velocity of this wave is as follows:

$$v_{\text{QL}}(\theta_{ac}) = \sqrt{\frac{[(C'_{11}(\varphi_Z) + C_{44})\cos^2\theta_{ac} + (C_{44} + C_{33})\sin^2(\theta_{ac})]}{2\rho} + \frac{\sqrt{[(C'_{11}(\varphi_Z) + C_{44})\cos^2\theta_{ac} - (C_{44} + C_{33})]^2\sin^2(\theta_{ac})} + (C_{13} + C_{44})\sin^2 2\theta_{ac}}{2\rho}}, \quad (8)$$

where

$$C'_{11}(\varphi_Z) = C_{11} - 0.5(C_{11} - C_{12} - C_{66})\sin 2\varphi_Z + C_{16}\sin 4\varphi_Z. \quad (9)$$

The orientation of the displacement vector of QL AW with respect to the wavevector is determined as:

$$\xi(\theta_{ac}, \varphi_Z) = 0.5 \text{atan} \left[ \frac{(C_{13} + C_{44})\sin 2\theta_{ac}}{(C'_{11}(\varphi_Z) - C_{44})\cos^2\theta_{ac} + (C_{44} - C_{33})\sin^2\theta_{ac}} \right]. \quad (10)$$

The effective EO coefficient can be written as:

$$p_{eff}^2(\theta_{ac}, \varphi_Z) = 0.5 \left\{ \begin{aligned} & p'_{61}(\varphi_Z) \cos^2 \theta_{ac} \cos^2 \xi + p_{55} \sin^2(\theta_{ac} + \xi) \\ & + (p'_{11}(\varphi_Z) \cos \theta_{ac} \cos \xi + p_{13} \sin \theta_{ac} \sin \xi)^2 \end{aligned} \right\} + 0.5 \left\{ \begin{aligned} & (p'_{21}(\varphi_Z) \cos \theta_{ac} \cos \xi + p_{23} \sin \theta_{ac} \sin \xi)^2 \\ & + p'_{61}(\varphi_Z) \cos^2 \theta_{ac} \cos^2 \xi + p_{45} \sin^2(\theta_{ac} + \xi) \end{aligned} \right\}, \quad (11)$$

where

$$p'_{61}(\varphi_Z) = 0.25(p_{12} - p_{11} + 2p_{66}) \sin 4\varphi_Z + (p_{16} + p_{61}) \cos^2 2\varphi_Z - p_{16}, \quad (12)$$

$$p'_{11}(\varphi_Z) = p_{11} - 0.5(p_{11} - p_{12} - 2p_{66}) \sin^2 2\varphi_Z + 0.5(p_{16} + p_{61}) \sin 4\varphi_Z, \quad (13)$$

$$p'_{21}(\varphi_Z) = 0.25(p_{11} + 3p_{12} - 2p_{66}) - 0.25(p_{11} - p_{12} - 2p_{66}) \cos 4\varphi_Z - 0.5(p_{16} + p_{61}) \sin 4\varphi_Z, \quad (14)$$

Now, let's consider the AO interaction with quasi-transverse QT<sub>2</sub> AW. The velocity of this AW does not depend on the angle  $\varphi_Z$  in the XY plane. Therefore, the only criterion for selecting the most suitable plane for AO interaction is the maximum value of the effective EO coefficient in that plane. However, the effective EO coefficient in the XZ plane is determined as:

$$p_{eff}^2 = 0.5(p_{55}^2 + p_{45}^2), \quad (15)$$

being independent of  $\varphi_Z$ . This value is equal to  $p_{eff}^2 = 3.61 \times 10^{-4}$ . Therefore, we will consider the AO diffraction in the XZ plane for this type of interaction.

The velocity of QT<sub>2</sub> AW is as follows:

$$v_{QT_2}(\theta_{ac}) = \sqrt{\frac{(C'_{11}(\varphi_Z) + C_{44}) \cos^2 \theta_{ac} + (C_{44} + C_{33}) \sin^2 \theta_{ac}}{2\rho}} \cdot \sqrt{\frac{((C'_{11}(\varphi_Z) + C_{44}) \cos^2 \theta_{ac} - (C_{44} + C_{33}) \sin^2 \theta_{ac})^2 + (C_{13} + C_{44}) \sin^2 2\theta_{ac}}{2\rho}}. \quad (16)$$

The effective EO coefficient can be written as

$$p_{eff}^2(\theta_{ac}, \varphi_Z) = 0.5 \left\{ \begin{aligned} & (p_{13} \sin \theta_{ac} \cos \xi - p'_{11}(\varphi_Z) \cos \theta_{ac} \sin \xi)^2 \\ & + p'_{61}(\varphi_Z) \cos^2 \theta_{ac} \sin^2 \xi + p_{55} \cos^2(\theta_{ac} + \xi) \end{aligned} \right\} + 0.5 \left\{ \begin{aligned} & (p_{23} \sin \theta_{ac} \cos \xi - p'_{21}(\varphi_Z) \cos \theta_{ac} \sin \xi)^2 \\ & + p'_{61}(\varphi_Z) \cos^2 \theta_{ac} \sin^2 \xi + p_{45} \cos^2(\theta_{ac} + \xi) \end{aligned} \right\}, \quad (17)$$

where angle  $\xi$  is given by Eq. (10) by adding 90 deg. Finally, the AO figure of merit is given by the formula:

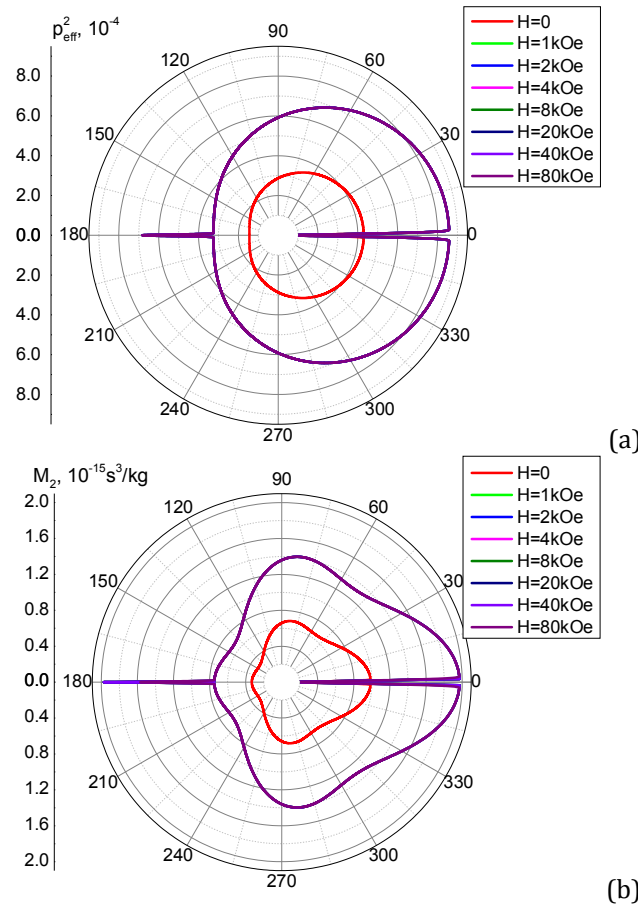
$$M_2 = \frac{n_i^3 n_d^3 p_{eff}^2}{\rho v^3}. \quad (18)$$

Here  $n_i$  and  $n_d$  denote the refractive indices of respectively incident and diffracted waves.

### 3. Results and discussion

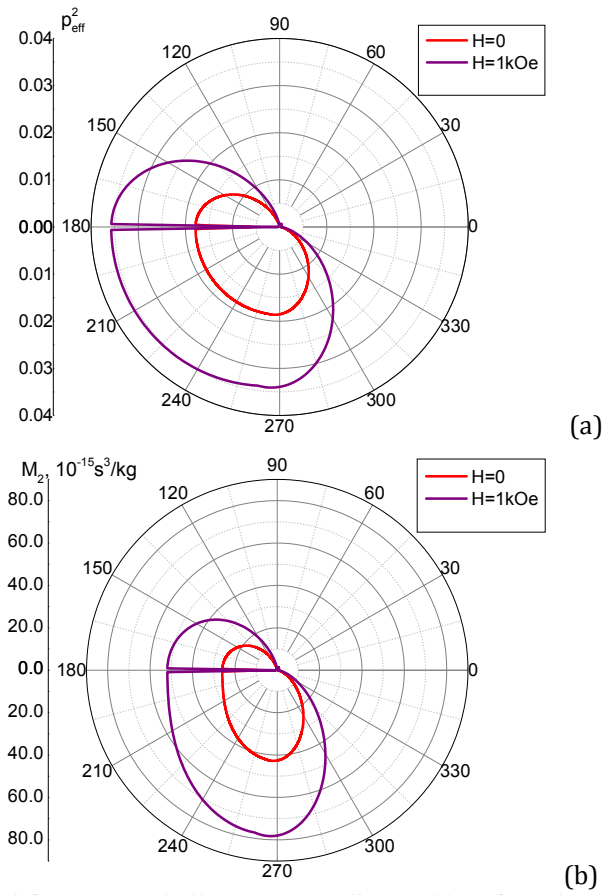
For the AO Bragg diffraction in the plane, which is rotated around the Z axis by 62 deg, the dependencies of the effective EO coefficient and the AO figure of merit are presented in Fig. 3a,b. It is seen that application of a magnetic field results in a jump-like increase in the effective EO coefficient and AO figure of merit. Further increasing the magnetic field does not alter the values mentioned above, as the ellipticity of the eigen optical waves becomes equal to  $\pm 1$  at a negligibly small magnitude of the magnetic field and remains constant with increasing field. It means that the effective EO coefficient and AO figure of merit acquire only two values – at switching off and

at the switching on magnetic field (see e.g. Eq. 3). Such behaviour is observed at the AO interaction with QT<sub>2</sub> and QL AWs (see below) therefore below for these types of AO interaction we will present dependencies for only one magnitude of magnetic field.



**Fig. 3.** Dependencies of the square of effective EO coefficient (a) and AO figure of merit (b) on the diffraction angle at the anisotropic AO interaction in the  $X'Z$  plane with QT<sub>1</sub> AW.

As one can see, the essential increase in the effective EO coefficient (Fig. 3a) and AO figure of merit is observed at all values of diffraction angle. For example, at the diffraction angle equal to 1 deg, the increase in the AO figure of merit is from 1 to  $2 \times 10^{-15} \text{ s}^3/\text{kg}$ . The exception is zero diffraction angle, which corresponds to the forward collinear AO diffraction. Notice that such diffraction cannot be realized at zero magnetic field, due to the lack of a phase-matching condition. In a non-zero magnetic field, forward collinear diffraction cannot be achieved because, to satisfy phase matching conditions, the wavelength of the AW must be at least an order of magnitude larger than the size of commonly used crystals. However, at a nonzero value of magnetic field, the backward collinear AO diffraction can be realized between the circularly polarized optical eigenwaves, which appear due to the circular birefringence induced by the Faraday effect. The AO figure of merit for such a kind of diffraction is equal to  $2 \times 10^{-15} \text{ s}^3/\text{kg}$ . At the switching off of the magnetic field, this diffraction vanishes. The frequency of AW that participates in the backward AO collinear diffraction is quite high and corresponds to the GHz frequency range. Although magnetic fields lead to an increase in the AO figure of merit at this type of anisotropic AO interaction, the magnitude of this parameter is comparatively small.



**Fig. 4.** Dependencies of the square of effective EO coefficient (a) and AO figure of merit (b) on the diffraction angle at the anisotropic AO interaction in the XZ plane with QT<sub>2</sub> AW.

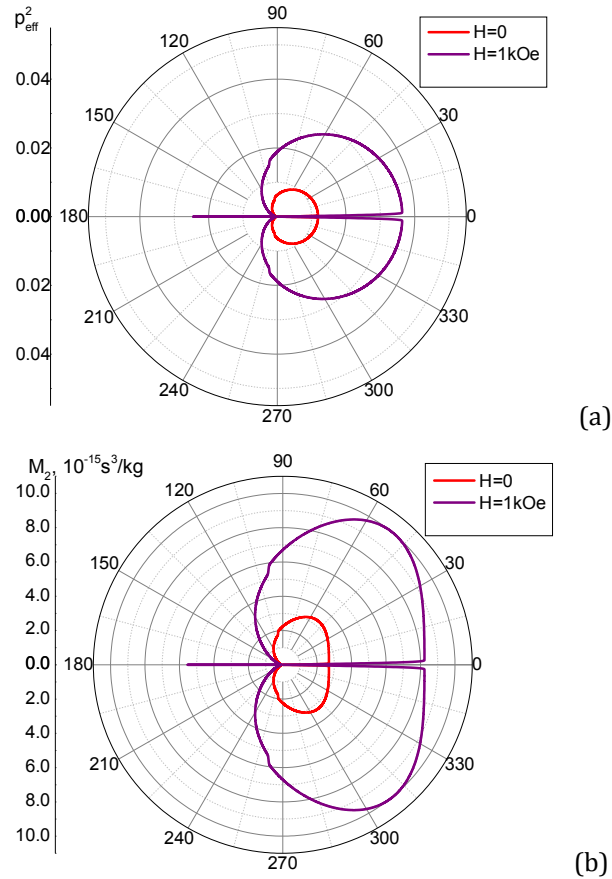
At the AO interaction with the QT<sub>2</sub> AW, the square of the effective EO coefficient and the AO figure of merit reach quite high values (Fig. 4). For example, the maximum AO figure of merit is 43 without a magnetic field applied and  $78 \times 10^{-15} \text{ s}^3/\text{kg}$  with the magnetic field turned on. However, this occurs at less convenient conditions, specifically at a diffraction angle of 267 deg (or -93 deg). At smaller diffraction angles, just a few degrees, the AO figure of merit does not exceed  $10^{-15} \text{ s}^3/\text{kg}$ .

The dependencies of effective EO coefficient and AO figure of merit on the diffraction angle for the AO interaction with the QL AW are presented in Fig. 5. The interaction plane is rotated by 17 deg around the Z axis. At the small angles of diffraction, the AO figure of merit changes from a value of 0.3 at zero magnetic field to  $8 \times 10^{-15} \text{ s}^3/\text{kg}$  at the switched-on magnetic field. Therefore, this type of AO interaction is most preferable from the perspective of experimental verification. The backward collinear diffraction is possible too at this type of AO interaction, i.e., one can see the peak of the effective EO coefficient and the AO figure of merit at  $\gamma=180$  deg.

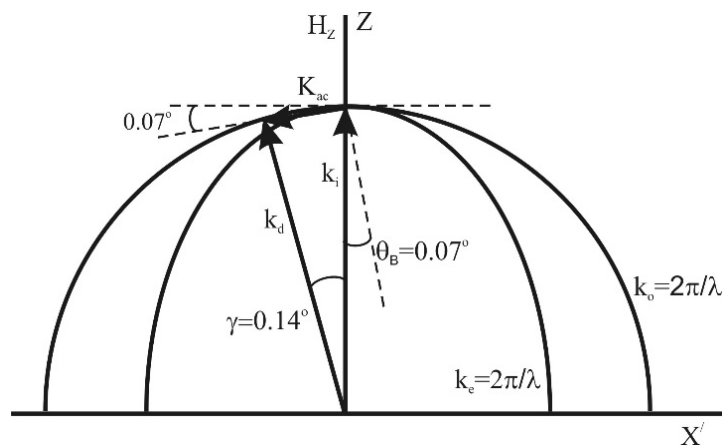
The actual geometry of the experiment slightly differs from the one described above for QL AW. The Bragg angle, which is calculated for the QL AW that propagates at a velocity of 5568 m/s, using the formula:

$$\sin \theta_B = \frac{\lambda}{2\Lambda_{ac}n_o}, \quad (19)$$

is equal to  $0.07^\circ$  (where AW wavelength  $\Lambda_{ac} = 111 \mu m$  at the AW frequency 50 MHz). Then the diffraction angle is two times greater than the Bragg angle, being equal to  $0.14^\circ$ . Therefore, the direction of propagation of the AW differs by  $0.07^\circ$  from  $X'$  direction (see Fig. 6). The oblique of the AW wave propagated in this direction is quite small, being equal to  $0.14^\circ$  [6].



**Fig. 5.** Dependencies of the square of effective EO coefficient (a) and AO figure of merit (b) on the diffraction angle at the anisotropic AO interaction in the  $X'Z$  plane with QLAW.



**Fig. 6.** Phase matching conditions for experimentally verifying anisotropic AO diffraction on the QL AW under a magnetic field.



#### 4. Conclusion

In the present work, we have considered the anisotropic AO diffraction in  $\text{NaBi}(\text{MoO}_4)_2$  crystals, taking into account the induction of circular birefringence by the magnetic field due to the Faraday effect. In geometry, when the incident optical wave propagates along the optical axis and the QL AW propagates almost parallel to the  $X'$  axis, which is turned on the  $X$  axis in the  $XY$  plane, the AO figure of merit reaches its highest value among other types of AO interactions with  $\text{QT}_1$  and  $\text{AT}_2$  AWs. Furthermore, the AO figure of merit changes from a value of  $0.3 \times 10^{-15} \text{ s}^3/\text{kg}$  at zero magnetic field to  $8 \times 10^{-15} \text{ s}^3/\text{kg}$  when the magnetic field is applied. We have proposed the actual experimental geometry for further validation of our theoretical predictions.

#### Funding

The study was supported by the Ministry of Education and Science of Ukraine (the research projects # 0123U101781 and 0125U002027).

#### Disclosures

The authors declare no conflicts of interest.

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**Анотація.** У цій роботі розглядається анізотропна акустооптична (АО) дифракція в кристалах  $\text{NaBi}(\text{MoO}_4)_2$  під впливом магнітного поля. Було показано, що індуковане магнітним полем циркулярне двопронезаломлення внаслідок ефекту Фарадея може призвести до підвищення ефективності АО дифракції, якщо падаюча оптична хвиля має кругову поляризацію та поширюється вздовж оптичної осі. Було показано, що при взаємодії з квазіпоздовжньою акустичною хвилею коефіцієнт АО якості змінюється від значення  $0,3 \times 10^{-15} \text{ с}^3/\text{кг}$  при нульовому магнітному полі до  $8 \times 10^{-15} \text{ с}^3/\text{кг}$  при прикладеному магнітному полі. Запропоновано геометрію для подальшої експериментальної перевірки наших теоретичних прогнозів.

**Ключові слова:** акустооптична якість, ефект Фарадея, магнітне поле, кристал подвійного молібдату натрію вісмуту