Anisotropy of acousto-optic figure of merit for the collinear diffraction of circularly polarized optical waves at the wavelength of isotropic point in AgGaS$_2$ crystals

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Abstract. We analyze anisotropy of acousto-optic figure of merit for a special case of collinear acousto-optic interactions between circularly polarized optical eigenwaves. These interactions take place in an optically active AgGaS$_2$ at the wavelength of a so-called isotropic point. The anisotropy of acoustic properties of the AgGaS$_2$ crystals is scrutinized and the cross sections of acoustic-wave velocity surfaces are obtained. We show that the acoustic walk-off angle reaches ~ 36 deg inside the crystallographic planes, while the non-orthogonality of polarization of the acoustic waves remains small and does not exceed ~ 7 deg in the principal planes. Theoretical relations for the effective elastooptic coefficients are obtained for the case of collinear diffraction. The maximal acousto-optic figure of merit, $0.9 \times 10^{15}$ s$^2$/kg, is reached when the interacting waves propagate at the angles 42, 138, 222 or 318 deg with respect to the a (b) axis in the ac (bc) plane. When the collinear acousto-optic diffraction is considered and the case of so-called acousto-gyration diffraction is disregarded, the angular momentum of circularly polarized photons can be transferred only into mechanical angular momentum of a crystalline sample.

Keywords: collinear acousto-optic diffraction, circularly polarized waves, isotropic point, optical activity, AgGaS$_2$ crystals

UDC: 535.42+535.56

1. Introduction

In our recent work [1] we have considered the acousto-optic (AO) interactions between circularly polarized optical eigenwaves. Circular waves can play a role of eigenwaves in an optically active medium whenever the linear birefringence is absent. Gases [2, 3], liquids [4], solid polymers [5], and crystals (see, e.g., Ref. [6]) are among those media which can reveal a natural optical activity and no linear birefringence. The appropriate conditions are cubic symmetry of the medium or propagation of light beam along one of the optic axes in optically uniaxial or biaxial crystals. For the media with cubic or higher symmetries the indicative surfaces of refractive indices for the left-handed (LH) and right-handed (RH) optical waves represent spheres of different radii, while in the optically uniaxial or biaxial crystals circular birefringence is hidden by linear birefringence, which is usually much larger than its circular counterpart. The circular birefringence caused by the optical activity is given by the relation (see, e.g., Ref. [7]):

$$\Delta n_c = \pm n_0^2 G, \ G = g_{\text{an}} l_l l_s.$$  \hspace{1cm} (1)

Here $l_1 = \sin \theta \cos \phi$, $l_2 = \sin \theta \sin \phi$ and $l_3 = \cos \theta$ are the components of the unit optical wave vector in the spherical coordinate system, with $\theta$ and $\phi$ denoting respectively the polar and azimuthal angles, $g_{\text{an}}$ the axial symmetric second-rank gyration tensor, $G$ the pseudo-scalar gyration parameter, and $n_0$ the ordinary refractive index.
Probably, a unique possibility for manifestation of anisotropy of the circular birefringence is a situation when the linear birefringence equals to zero at the wavelength of isotropic point $\lambda_i$ in non-cubic crystals. Then the condition of equality of the ordinary and extraordinary refractive indices is satisfied. Under this condition, splitting of the indicative surfaces of the refractive indices for the LH ($n_o$) and RH ($n_e$) waves is expected. The appropriate relation is as follows:

$$n_{o,e} = n_s \pm \frac{1}{2} n_s^2 G$$  \hspace{1cm} (2)

Since the $G$ parameter depends on the constitutive gyration tensor, the refractive-index surfaces for the LH and RH waves in the crystals belonging to middle- or lower-symmetry systems are no longer spheres but represent complicated high-order surfaces \[8,\ 9\].

Under such conditions, one can anticipate AO interactions between the circular optical eigenwaves and, in particular, an anisotropic Bragg diffraction at which the incident and diffracted optical waves have the opposite circular polarizations. In our recent work \[1\], we have considered three particular cases of AO interactions between the circular optical waves in AgGaS$_2$ (the point symmetry group \$42m\ [10]\). These are isotropic, anisotropic and collinear anisotropic diffractions at the wavelength of the isotropic point ($\lambda_i = 497.4$ nm and $n_s = 2.685$ \[10\]). It has been found in Ref. \[1\] that the AO figure of merit (AOFM) defined as

$$M_2 = n_s^6 p_{ij}^2 / \rho v_i^3$$  \hspace{1cm} (3)

is quite small at the chosen geometry of anisotropic interactions. We remind that, in Eq. (3), $p_{ij}$ is the effective elasto-optic (EO) coefficient, $v_i$ the acoustic wave (AW) velocity (with $i$ and $j$ corresponding respectively to the directions of AW propagation and polarization), and $\rho$ the crystal density. In particular, the AO figure of merit does not exceed $\sim 10^{-15}$ s$^3$/kg at the collinear diffraction of the waves that propagate at the angle 45 deg with respect to the $a$ axis in the crystallographic plane $ac$. Hence, it would be reasonable to study the anisotropy of AO figure of merit in order to find more efficient geometries of the interactions. We stress in this respect that only modules of the EO coefficients have been determined in Ref. \[1\], although the signs of all the EO coefficients are usually needed to determine the effective EO coefficients. The only exception is concerned with the AO interactions inside the crystallographic planes, where the effective EO coefficient is given by a single EO tensor component.

In the present work we obtain phenomenological relations for the effective EO coefficients at the collinear interactions of circularly polarized optical waves with acoustic eigenwaves in the AgGaS$_2$ crystals. Besides, we analyze the anisotropy of AOFM inside the crystallographic planes.

2. Results and their discussion
Let us analyze the anisotropy of AW velocities in the AgGaS$_2$ crystals. For this aim one needs the elastic stiffness coefficients, which have been determined in Ref. \[11\] as follows: $C_{11} = 8.79 \pm 0.05$, $C_{12} = 7.58 \pm 0.05$, $C_{44} = 2.41 \pm 0.05$, $C_{66} = 3.08 \pm 0.05$, $C_{12} = 5.84 \pm 0.05$, and $C_{13} = 5.92 \pm 0.06$ (in the units of 10$^{10}$ N/m$^2$). Using the Christoffel equation \[12\] and the density $\rho = 4700$ kg/m$^3$ of AgGaS$_2$ \[11\], we have obtained cross sections of AW velocity surfaces by the crystallographic planes $ac$ and $ab$ (see Fig. 1a, b). One can see that the velocities of quasi-transverse (QT) AWs with orthogonal polarisations are equal along the directions of ‘acoustic axes’. One of these axes (AA$_1$) is parallel to the $c$ axis (see Fig. 1a) and four axes lie in the $ab$ plane (see Fig. 1b). The angles between the latter axes and the $a$ axis are equal to 72 deg for the AA$_2$ axis, 18 deg for AA$_3$, -18 deg for AA$_4$ and -72 deg for AA$_5$.
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A schematic view of a part of the surface of velocities for the acoustic eigenwaves and their polarizations are presented in Fig. 1c in a single octant of coordinate system. The points of outlets of the acoustic axes $AA_i$–$AA_j$ represent polarization topological defects with the strength $\frac{1}{2}$. There occurs switching of the polarization by 90 deg under the condition when one passes through a defect on the QT velocity surface (see Fig. 1c). Here the outlet of the acoustic axis $AA_i$ represents the polarization topological defect with the strength equal to unity.

The obliquity angle between the acoustic group-velocity direction and the AW vector has been calculated using the relation [13]

$$\Delta_i = \arctan \frac{1}{v(\Theta_i)} \frac{\partial v(\Theta_i)}{\partial \Theta_i}.$$  \hspace{1cm} (4)

Here $v(\Theta_i)$ denotes a function of AW velocity that depends upon the angle $\Theta_i$ between the wave vector and the corresponding axis of the crystallographic coordinate system, with the subscript $i$ referring to the axis perpendicular to the geometric plane under consideration. As seen from Fig. 2a and Fig. 2c, the walk-off angle reaches high values for the QT waves, 39 and 36 deg respectively in the $ac$ ($bc$) and $ab$ planes. The walk-off angle for the quasi-longitudinal (QL) wave is smaller. It achieves its maximal value equal to 5 and 10 deg in the $ac$ ($bc$) and $ab$ planes, respectively.

The angle of deviation of the AW polarization from the purely longitudinal types has been calculated basing on the Christoffel equation [14]:

$$\zeta_{1,2} = \frac{1}{2} \arctan \frac{(C_{11} + C_{44}) \sin 2\Theta_{1,2}}{(C_{11} - C_{44}) \cos^2 \Theta_{1,2} + (C_{44} - C_{13}) \sin^2 \Theta_{1,2}},$$ \hspace{1cm} (5)

$$\zeta_3 = \frac{1}{2} \arctan \frac{(C_{11} + C_{66}) \sin 2\Theta_3}{(C_{11} - C_{66}) \cos^2 \Theta_3 + (C_{66} - C_{11}) \sin^2 \Theta_3}. \hspace{1cm} (6)$$

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Dependences of the angle of non-orthogonality of AW polarizations on the direction of AW propagation are presented in Fig. 2b and Fig. 2c. The angle of deviation of the AW polarizations from the purely longitudinal and transverse states remains small and does not exceed ~7 deg. Therefore one can neglect the above angle in the further analysis.

![Graph](image1)

**Fig. 2.** Obliquity angle (a, c) of AWs and angle of non-orthogonality of AW polarizations (b, d) in the ac (bc) (a, b) and ab (c, d) planes as functions of angle $\Theta$, between the wave vector direction and the a axis.

Using the approaches introduced in Refs. [1, 15, 16], we have derived the effective EO coefficients for the collinear AO interactions of circular optical waves with the acoustic eigenwaves under the condition when the non-orthogonality of their polarization is neglected:

\[
p_{\text{eff}}^{QL} = -0.25 \cos 2\varphi \sin 2\varphi \sin \theta \cos^2 \theta (2p_{21} - 2p_{11} + p_{bb}),
\]

\[
p_{\text{eff}}^{QT_1} = -0.5 \cos 2\varphi \sin 2\varphi \cos \theta \sin^2 \theta (2p_{21} - 2p_{11} + p_{bb}),
\]

\[
p_{\text{eff}}^{QT_2} = 0.5 \sin 2\theta \left( \frac{D_{ab} \cos 2\varphi - p_{ab}}{2} + \sin^2 2\varphi (p_{11} - p_{21}) \right).
\]

It is seen from Eqs. (7)–(9) that the effective coefficients depend on a number of EO coefficients, the signs of which are not determined yet. In the principal crystallographic plane ab ($\theta = 90$ deg), all the effective coefficients are equal to zero, thus making the AO interactions impossible. The AO interactions with the AWs QL and QT$_1$ are impossible in the ac (bc) plane at $\varphi = 0$ or 90 deg due to the same reason. However, the collinear AO interactions with the AW labelled as QT$_2$ according standard notation can be implemented at $\varphi = 0$ or 90 deg. In this case Eq. (9) can be rewritten as
$$p_{\theta}^{(0)} = \frac{p_{\theta} - p_{\theta}}{4} \sin 2\theta . \tag{10}$$

With taking the equalities $p_{\theta} \approx 0$ and $p_{\theta} = 0.053$ into account (see Ref. [1]), one can simplify Eq. (10) to the form $p_{\theta}^{(0)} = \frac{1}{4} p_{\theta} \sin 2\theta$. The dependence of the relevant effective EO coefficient on the angle $\theta$ is displayed in Fig. 3a. The maximal value of the effective coefficient is reached whenever the interacting waves propagate at the angles 45, 135, 225 or 315 deg with respect to the $a$ ($b$) axis in the $ac$ ($bc$) plane. The anisotropy of the inverse cube of velocity for the AW QT$_2$ in these planes (see Fig. 3b) is not notably pronounced. Thus, the AOFM anisotropy should be mainly caused by the anisotropy of effective EO coefficients. As a result, the AOFM reaches it maximal value equal to $0.9 \times 10^{-15}$ s$^3$/kg at the angles $\theta = 42, 138, 222$ or 318 deg. A difference of ±3 deg between the angles of maximal AOFMs and the angles of maximal effective EO coefficients is caused by the anisotropy of QT$_2$ wave velocity.

![Effective EO coefficient (a), inverse cube of the QT$_2$ velocity (b) and AOFM (c) in the ac (ab) plane as functions of the polar angle $\theta$.](image)

Fig. 4 shows dependence of the AW frequency $f_A$ that satisfies the phase matching condition $k_s + k_{ac} = k_l$ (with $k_s$ and $k_l$ being the wave vectors of the optical RH and LH waves, and $K_{ac}$ denoting the AW vector) on the polar angle $\theta$. This dependence concerns the collinear AO diffraction of circularly polarized optical waves at the AW QT$_2$. The $f_A$ frequency changes from zero up to 7.4 MHz when the propagation direction for the interacting waves changes from $\theta = 90$ deg (i.e., along the $a$ ($b$) axis) to $\theta = 0$ deg (i.e., along the $c$ axis). It should be noted that the AO interactions cannot be implemented at $\theta = 0$ deg because of the absence of circular birefringence. Moreover, the effective EO coefficient is equal to zero at $\theta = 90$ deg.
(i.e., in the $ab$ plane), thus making impossible the collinear AO interactions. Finally, the frequency of AW is equal to 3.1 MHz at $\theta = 42$ deg, which corresponds to the AOFM maximum.

The spin angular momentum of the interacting photons changes as $h \rightarrow -h$ at the collinear anisotropic AO interactions between the incident optical wave RH and the diffracted LH wave. The conservation law for the angular momentum requires the condition $h = -h + M$, where $M = 2h$ is the orbital angular momentum of the diffracted beam [17] or the mechanical angular momentum transferred to a crystalline sample. Notice that the axial symmetry of the collinear AO interactions can be preserved only in the case of interaction with the longitudinal AW propagating along the three-, four- or six-fold symmetry axes. In this case the AW induces no birefringence along the direction of light propagation. However, since the above symmetry axes are parallel to the optic axes in the uniaxial crystals, the AO interactions with the longitudinal AWs cannot be implemented along these directions. The reason is zero values of the corresponding elasto-optic (EO) coefficients. The same is true of the optically active crystals of cubic system when the interacting waves propagate along the three-fold symmetry axes. Hence, the angular momentum of circularly polarized photons at the collinear AO diffraction can be transferred only into the mechanical angular momentum of a crystalline plate, quite similar to the Beth’s effect [18]. This statement does not refer to the case of acousto-gyration diffraction (see Refs. [19, 20]).

3. Conclusions
We have analyzed the anisotropy of acoustic and AO properties of the AgGaS$_2$ crystals. It has been found that these crystals have five ‘acoustic axes’. The velocities of the QT AWs are equal to each other whenever these waves propagate along the above axes. One of the acoustic axes is parallel to the $c$ axis and the four remaining ones lie in the crystallographic plane $ab$. The obliquity of the acoustic energy flow with respect to the AW vector is high enough for the AgGaS$_2$ crystals. For instance, the walk-off angle reaches the value $\sim 36$ deg inside the crystallographic planes. At the same time, the angle of non-orthogonality of AW polarizations is small and does not exceed $\sim 7$ deg inside the principal planes. This allows one to carry out the analysis of AOFM anisotropy in the approximation when the non-orthogonality effect is disregarded.

The AO properties of AgGaS$_2$ have been analyzed for the special case of collinear AO diffraction of circular optical waves with the wavelength $\lambda = 497.4$ nm that correspond to the ‘isotropic point’. The relations for the effective EO coefficients for the collinear AO diffraction have been obtained. We have found that the anisotropy of AOFM in the crystallographic planes $ac$ and $bc$ is mainly caused by the anisotropy of effective EO coefficient. The maximal AOFM value,
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0.9×10^{-15} \text{s}^3/\text{kg}, is reached in case if the interacting waves propagate at the angles $\theta = 42$, 138, 222 or 318 deg in the ac (bc) plane. Finally, we have shown that, at the collinear AO diffraction, the angular momentum of circularly polarized photons can be transferred only into the mechanical angular momentum of a crystalline sample. This is not valid for the case of so-called acoustogyration diffraction.

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Anotacія. Проаналізовано анізотропію коефіцієнта акустооптичної якості для колінеарних акустооптичних взаємодій між циркулярно поляризованими циркулярно поляризованими останніми хвилями. Ці взаємодії мають місце в оптично активних кристалах AgGaS$_2$ на довжині хвилі так званої ізотропної точки. Досліджено анізотропію акустичних властивостей AgGaS$_2$ i одержано перерізи поверхонь швидкостей акустичних хвиль. Показано, що кут акустичного зносу в кристалографічних площинах досягає ~ 36 град, а неортогональність поляризації акустичних хвиль залишається малою і не перевищує ~ 7 град. Одержано теоретичні співвідношення для ефективних пружнооптичних коефіцієнтів для випадку колінеарної дифракції. Максимальна коефіцієнта акустооптичної якості (0,9×10$^{-15}$ с$^3$/кг) досягаємо, коли взаємодіючі хвилі поширюються під кутами 42, 138, 222 або 318 град до осі a (b) у площинах ac (bc). Якщо розглядати колінеарну акустооптичну дифракцію та виключити випадок так званої акустогіраційної дифракції, то момент імпульсу циркулярно поляризованих фотонів можна перетворити тільки на механічний момент імпульсу кристалічного зразка.

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Анотація. Проаналізовано анізотропію коефіцієнта акустооптичної якості для колінеарних акустооптичних взаємодій між циркулярно поляризованими хвилами. Ці взаємодії мають місце в оптично активних кристалах AgGaS$_2$ на довжині хвилі так званої ізотропної точки. Досліджено анізотропію акустичних властивостей AgGaS$_2$ і одержано перерізи поверхонь швидкостей акустичних хвиль. Показано, що кут акустичного зносу в кристалографічних площинах досягає ~ 36 град, а неортогональність поляризації акустичних хвиль залишається малою і не перевищує ~ 7 град. Одержано теоретичні співвідношення для ефективних пружнооптичних коефіцієнтів для випадку колінеарної дифракції. Максимальна коефіцієнта акустооптичної якості (0,9×10$^{-15}$ с$^3$/кг) досягаємо, коли взаємодіючі хвилі поширюються під кутами 42, 138, 222 або 318 град до осі a (b) у площинах ac (bc). Якщо розглядати колінеарну акустооптичну дифракцію та виключити випадок так званої акустогіраційної дифракції, то момент імпульсу циркулярно поляризованих фотонів можна перетворити тільки на механічний момент імпульсу кристалічного зразка.