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# Research of Optical Activity and Circular Dichroism in Some Uniaxial Crystals in Directions Different from an Optical Axis

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## Abstract

The research of optical activity in some uniaxial crystals was done based on the spectropolarimeter method. The measurements were conducted on the plates cut perpendicularly to and parallel to axis in visible spectrum range. Dispersion of all components of gyration pseudotensor for a number of uniaxial crystals was determined for the first time from these measurements. The absorption spectra and circular dichroism of crystals of gallogermanate  $\text{Sr}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}$ , undoped and doped by chromium were investigated.

## 1. Introduction

The special place among optical properties of crystals occupies a phenomenon of optical activity, or gyrotropy [1,2]. Two circular polarized waves propagate in absorptive isotropic media and in anisotropic crystals in the direction of optical axis. That results in rotation of the plane of polarization and circular dichroism. In directions different from optical axis two elliptically polarized waves propagate. In that case it is possible to talk about elliptical birefringence and elliptical dichroism. The optical activity is described by a gyration pseudotensor of the second rank. (a prefix pseudo we will omit). By measuring rotation of the plane of polarization it is possible to determine only one component. Other components can be determined if measurements are conducted on plates cut in directions different from optical axis. That is not a simple problem, because it is necessary to determine small components of a gyration tensor on a background of rather large birefringence. Therefore to the present time very few crystals are known, for which all components of a gyration tensor are determined even though these values are relevant characteristic of crystals.

The purpose of the present article is the research of an optical activity and also determination of components of a gyration tensor in some uniaxial crystals of a different symmetry class in directions different from direction from optical axis.

## 2. Theoretical Relations

All values describing optical properties of crystals are complex in absorptive gyrotropic crystals:

$$N_{1,2} = n_{1,2} + i\kappa_{1,2}, G = G' + iG'', k = k' + ik'', \quad (1)$$

where  $N_1, N_2$ - complex refractive indices,  $n_1, n_2$  - refractive indices,  $\kappa_1, \kappa_2$  - absorption coefficients,  $G$  - scalar parameter gyration, which is determined by components of a gyration tensor and depends on a symmetry of crystal and propagation direction of light in crystal, ellipticity of eigen waves

propagating in crystal ( $b/a = \text{tg}\gamma$ ) and angle of non-orthogonality ( $\beta$ ) are determined from relation [2,3]:

$$\sin 2\gamma = 2k'/(1+k'^2+k''^2), \text{tg} 2\beta = 2k''/(1-k'^2-k''^2). \quad (2)$$

In transparent crystals ellipticity of eigen waves is equal  $k = k'$ .

In uniaxial crystals in direction parallel to optical axis eigen waves are circular polarized and we have

$$k' = \pm 1, k'' = 0, G = g'_{33} + ig''_{33}. \quad (3)$$

Component of gyration tensor  $g'_{33}$  is calculated from measurement of an angle  $\rho$  of rotation of polarization plane on a spectropolarimeter:  $g'_{33} = \rho \lambda n_0 / \pi$ . Component  $g''_{33}$  is calculated from a relation  $g''_{33} = \Delta \varepsilon \lambda n_0 / 4\pi c$ , where,  $\Delta \varepsilon = \Delta D/d$ ,  $\Delta D$  is optical density recorded on dichrograph,  $c$  is concentration of dope,  $d$  is thickness of investigated plate,  $\lambda$  is wavelength of incident light.

In directions perpendicular to direction of optical axis in uniaxial crystals  $G = g'_{11} + ig''_{11}$ , the values  $k'$  and  $k''$  are small and they are equal [3]:

$$k' = [g'_{11} \delta n_1 + g''_{11} \delta \kappa_1] / [2 n_0 (\delta n_1^2 + \delta \kappa_1^2)], \quad (4a)$$

$$k'' = [g''_{11} \delta n_1 - g'_{11} \delta \kappa_1] / [2 n_0 (\delta n_1^2 + \delta \kappa_1^2)], \quad (4b)$$

where  $\delta n_1 = (n_2 - n_1)$  is linear birefringence,  $\delta \kappa_1 = (\kappa_2 - \kappa_1)$  is linear dichroism,  $\delta n_c = g'_{11}/n_0$  is a circular birefringence,  $\delta \kappa_c = g''_{11}/n_0$  is a circular dichroism,  $n_0$  is an average value of refractive indexes.

In directions, different from optical axis we can obtain the most informative expressions for the azimuth  $\chi$  of passed light in cases, when the incident wave with azimuth  $\alpha$  is linearly polarized in direction parallel to ( $\parallel$ ) ( $\alpha = 0^\circ$ ) or perpendicular to ( $\perp$ ) ( $\alpha = 90^\circ$ ) principal plane (that is plane, in which optical axis and normal to the surface are located). Unlike inactive crystal in these cases the light propagated through a plate is not linear but elliptically polarized with azimuth  $\chi$ . When  $k'$  and  $k''$  are small we have for azimuth of transmitted light  $\chi_{\parallel, \perp}$ :

$$\text{tg} 2\chi_{\parallel, \perp} = -2 [e^{\pm \delta} k' \sin \Delta \pm k'' (1 + e^{\pm \delta} \cos \Delta)], \quad (5)$$

where  $\Delta = 2\pi d \delta n_1 / \lambda$ ,  $\delta = 2\pi d \delta \kappa_1 / \lambda$ .

The values of  $\text{tg} 2\chi_{\perp} \neq \text{tg} 2\chi_{\parallel}$  for different a wavelength of an incident light and they oscillate, since the periodic functions  $\sin \Delta$  and  $\cos \Delta$  change. It is possible to measure the azimuth of transmitted light  $\chi_{\parallel, \perp}$  on a spectropolarimeter for different wavelengths of an incident light. Thus it is necessary to know values of a birefringence and dichroism, which one can be calculate from the same experiment. In most cases  $k' > k''$ , then it is possible to consider that

$$\text{tg} 2\chi_{\parallel, \perp} = -2 e^{\pm \delta} k' \sin \Delta. \quad (6)$$

In transparent crystals the value  $\delta = 0$ . Then expression (5) becomes simpler. In these cases envelope of a curve of oscillating functions  $\chi_{\parallel} = \chi_{\perp}$  and we can determine the value  $k'$  and accordingly to calculate component  $g'_{11} = 2k' n_0 \delta n_1$ .

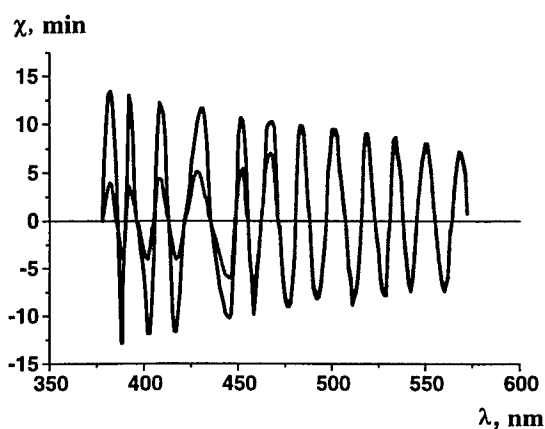
### 3. Determination of Components of Gyration Tensor in Uniaxial Crystals

We conduct the research of an optical activity of the following crystals: undoped and Cr-doped  $\text{Sr}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}$  and also  $\text{KH}_2\text{PO}_4$  (KDP),  $\text{NH}_4\text{H}_2\text{PO}_4$  (ADP),  $\text{K}_2\text{S}_2\text{O}_6$ ,  $\text{SrS}_2\text{O}_6 \cdot 4\text{H}_2\text{O}$ .

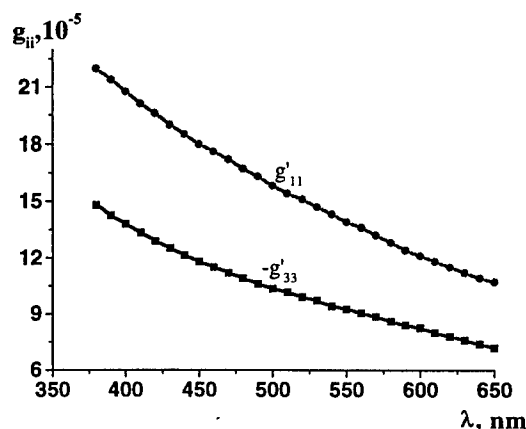
The crystals  $\text{Sr}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}$ ,  $\text{K}_2\text{S}_2\text{O}_6$ ,  $\text{SrS}_2\text{O}_6 \cdot 4\text{H}_2\text{O}$  and  $\text{Sr}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}$  have symmetry classes 32, 3 and 6 accordingly. Crystals of these classes of symmetry can rotate plane of polarization. The

investigated plates were cut parallel to and perpendicular to the  $c$ -axis of crystals with dimensions  $10 \times 10 \text{ mm}^2$  with thickness varying from 0.5 to 1.5 mm.

We conduct measurements of relations  $\chi_{\parallel, \perp}(\lambda)$  for all investigated plates on spectropolarimeter [4] using method, described in [3]. The dependence of azimuth  $\chi_{\parallel, \perp}$  for crystal  $\text{Sr}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}:\text{Cr}$  is shown on Fig. 1. According to relation (6), enveloped curve of oscillations is determined by  $e^{\pm\delta} k'$ . After determination of dispersion of an ellipticity of eigen waves  $k'$  and value  $\delta$  it is possible to calculate dispersion of components of a gyration tensor  $g'_{11}$ . In order to calculate the value  $g'_{11}$  except for measured values  $k'$  it is necessary to know values of birefringence, average refractive indices and value  $\delta$ . Values  $g'_{33}$  and  $g'_{11}$  for crystal  $\text{Sr}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}$  are shown in Fig. 2.



**Fig. 1** Dependence of azimuth of transmitted light  $\chi$  from wavelength  $\lambda$  for a plate cut from a crystal  $\text{Sr}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}:\text{Cr}^{3+}$  parallel to optical axis for two polarization of incident light ( $d = 0.96 \text{ mm}$ )



**Fig. 2** A dispersion of components of gyration pseudotensor  $g'_{11}$  and  $g'_{33}$  for crystal  $\text{Sr}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}:\text{Cr}^{3+}$

Absorption spectra for crystals  $\text{Sr}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}:\text{Cr}$  were measured in the range 250 - 800 nm. In absorption spectra of these crystals three bands connected with ions  $\text{Cr}^{3+}$  in octahedral coordination are found. The circular dichroism (CD) spectra were studied in the range 250 - 800 nm on dichrograph, constructed and made in Institute of Crystallography of RAS [5]. In order to calculate components of  $g'_{33}$  correctly, it is necessary to take into account concentration  $c$  of doped Cr. Since value  $c$  is not determined precisely, we present only dependence  $\Delta\epsilon$  of crystal  $\text{Sr}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}:\text{Cr}$  on Fig. 3. It is found that CD is present in all three broad absorption bands of spectra and in the narrow R-line ( $\sim 700\text{nm}$ ) of  $\text{Cr}^{3+}$  ions. In spectra of circular dichroism of doped crystals the bands of absorptions connected with ions  $\text{Cr}^{3+}$  were also found. Besides that in spectra of circular dichroism the additional bands are observed. These bands are not associated with ions  $\text{Cr}^{3+}$ . In spectra circular dichroism of undoped crystals the same bands CD were also found (curve 2). Probably all bands observed in the spectra of circular dichroism of undoped crystals can be attributed to defects generated during crystal growth.

Crystals KDP, ADP and  $\text{K}_2\text{S}_2\text{O}_6$ ,  $\text{SrS}_2\text{O}_6 \cdot 4\text{H}_2\text{O}$  were transparent in visible range of spectra. The crystals KDP and ADP belong to a symmetry class  $\bar{4}2m$  and do not have optical activity in the direction optical axis of a crystal. However, in crystals of given symmetry class the gyrotropy can appear in directions different from optical axis. In order to determine the components of gyration tensor the plates were cut in crystallographic directions X and Y.

Dependences of azimuth  $\chi_{\parallel, \perp}$  were obtained for these crystals. Now we have  $\chi_{\parallel} = \chi_{\perp}$ . From these dependences we calculated dispersion of components  $g'_{11}$ , which is shown in Fig. 4. Research was done for KDP crystals and ADP crystals on plates X and Y - cut of identical thickness. IN this case

azimuths  $\chi(\lambda)$  have opposite phases. Therefore  $g'_{11} = -g'_{22}$ . This is how it should be in crystals of this symmetry class. The value of component of gyration tensor  $g'_{11}$  for KDP crystal is larger than in ADP crystal.

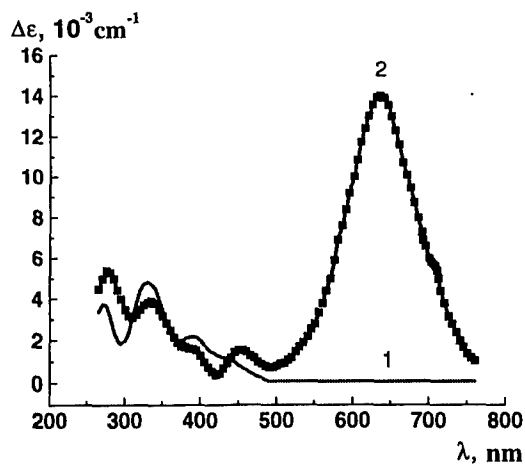


Fig. 3 Circular dichroism spectra of undoped crystal  $\text{Sr}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}$ : (curve 1) and Cr-doped crystal  $\text{Sr}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}$  (curve 2)

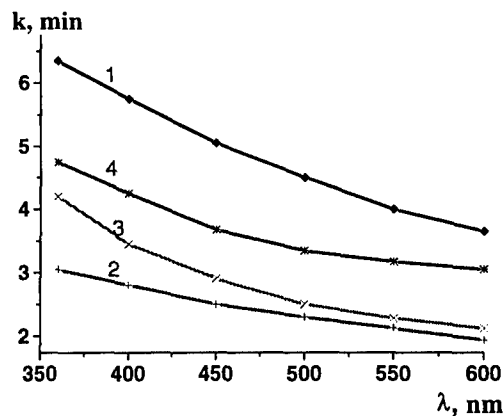


Fig. 4 A dispersion of components of gyration pseudotensor  $g'_{11}$  for crystals: 1-  $\text{KH}_2\text{PO}_4$  (KDP), 2-  $\text{NH}_4\text{H}_2\text{PO}_4$  (ADP) 3 -  $\text{K}_2\text{S}_2\text{O}_6$ , 4 -  $\text{SrS}_2\text{O}_6 \cdot 4\text{H}_2\text{O}$

The component  $g'_{11}$  for KDP crystal was determined in [6] for two wavelengths  $\lambda = 462 \text{ nm}$  and  $506 \text{ nm}$ . The values  $g'_{11}$ , obtained in this article is larger than these obtained by us. At the same time in more recent article of the same authors [7] values of  $g'_{11}$  are much less than values obtained by them in [6] and values obtained by us.

Dispersion of gyration tensor components  $g_{11}$  is obtained for crystals  $\text{Sr}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}$ , KDP, ADP,  $\text{K}_2\text{S}_2\text{O}_6$ ,  $\text{SrS}_2\text{O}_6 \cdot 4\text{H}_2\text{O}$  for the first time.

The absorption spectra and circular dichroism of gallogermanate crystals  $\text{Sr}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}$ , undoped and Cr-doped are investigated.

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