# Polarimetric Studies of Linear Dichroism in Cr-Doped Gallogermanate Crystals

Y. Shopa and N. Ftomyn

Ivan Franko National University of Lviv, 8 Kyrylo and Mefodiy St., 79005 Lviv, Ukraine

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## Abstract

A technique for simultaneous measurements of optical birefringence (OB) and linear dichroism (LD) with a high-accuracy computerized polarimeter is developed and applied to $Ca_3Ga_2Ge_4O_{14}:Cr^{3+}$ crystals. A characteristic behaviour of equi-intensity ellipses on the HAUP maps is found out. Temperature dependences of the OB changes and the LD are obtained for the temperature interval 295–353 K.

**Keywords**: optical birefringence, dichroism, polarimeter, HAUP map

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## 1. Introduction

Linear dichroism (LD) is a difference between the principal extinction coefficients of crystals. Serious experimental difficulties usually exist if one measures simultaneously different optical anisotropy parameters in optically birefringent crystals. This is because the linear optical birefringence (OB) is typically much larger than all the other effects. The latter situation occurs whenever the measurements are performed for the light propagation directions different from the optic axes. A number of methods have been suggested in the works for simultaneous measurements of optical anisotropy parameters of crystals in the directions that differ from the optic axes [1, 2]. Quantitative parameters of the LD have been obtained in these studies on the basis of analysis of azimuth oscillations for the light passed from a crystal, performed for different values of phase difference between the normal light waves.

On the other hand, high-accuracy polarimetric methods represent the most precise and convenient techniques for solving the mentioned crystal optical problems. In frame of the HAUP approach (the commonly adopted abbreviation HAUP means ‘high-accuracy universal polarimeter’), the optical transmission of the PSA system (P being a polarizer, S specimen and A an analyser) is determined as a function of $\theta$ and $Y$ azimuths [3–5]:

$$I = I_0\left(A(\theta) + B(\theta)Y + CY^2\right), \qquad (1)$$

where $I$ is the intensity of the emergent light, $I_0$ the intensity of the incident light, $\theta$ the polarizer azimuth, $Y$ the angle that defines deviation of the analyzer from the crossed-polarizers position and, finally, $A(\theta)$, $B(\theta)$ and $C$ represent functions of the optical anisotropy parameters. The OB and the LD (if a crystal is dichroic) might be obtained, provided that the dependences $A(\theta)$ and $B(\theta)$ are thoroughly analyzed [3, 5]. The main parameters referred to the PSA system in case of dichroic crystals have been introduced in the studies [3, 4], where the authors have used the theoretical approach valid for extremely small dichroisms. An extended HAUP theory, which could be applied to crystals with arbitrary LD values, has been developed in the work [5]. The LD and the others parameters of optical anisotropy of crystals have been investigated by us in the work [6], using both the PSA and the PSCA systems (with C implying a compensator), including the cases of light propagation in crystal both along and perpendicular to the optic axes. However, the method suggested in [6] requires complicated measuring procedures and equipment. In the present paper we extend our polarimetric method for measuring LD in crystals for the propagation directions different from those coinciding with the optic axis. The method is based on the analysis of so-called HAUP maps and, moreover, it seems to be simpler for practical realization, when compare to the techniques mentioned above.

When examining our experimental technique, we have chosen $Ca_3Ga_2Ge_4O_{14}$ :$Cr^{3+}$ crystals as the objects under test, since they are optically active (the symmetry class 32) and possess a significant OB and some LD. These crystals are optically uniaxial and positive (the refractive indices of extraordinary ($n_e$) and ordinary ($n_o$) waves are related according to $n_e > n_o$) [7]. Notice that some experimental results already available for $Sr_3Ga_2Ge_4O_{14}$ :$Cr^{3+}$ crystals, which have been published in the work [2], have been got with a quite another method.

## 2. Experimental details

The optical scheme of our high-accuracy computerized polarimeter [8, 9] includes a minimal quantity of optical components: a polarizer, specimen and an analyser, thus forming the PSA system. He-Ne laser with the wavelength of $\lambda = 632.8$ nm has been used as a light source. Three characteristic azimuth angles of polarizer have been measured during the experiment. The corresponding measuring procedures employed for this aim have been earlier described in [9]. An extra feature of our present technique is a detailed analysis of the so-called HAUP maps. As a consequence, this technique is different from the HAUP one, though we calculate the transmission of the PSA system, too.

The transmission $J$ of the PSA system is a function of the two azimuths, those of the polarizer ($\theta$) and the analyzer ($\chi$), which are measured from the principal optical ellipsoid axes. Within the limits of small changes in $\theta$ and $\chi$ angles (less than 0.01 rad), the relative intensity $J(\theta,\chi) = I/I_0$ for dichroic crystals may be expressed as

$$\begin{gathered} J(\theta,\chi) = e^{-E}\theta^2 + e^{E}\chi^2 - 2\theta\chi\cos\Gamma + \\ + f_1\theta + f_2\chi + \text{const} \end{gathered}, \quad (2)$$

where $E = \frac{2\pi}{\lambda}\left(m_z - m_y\right)d$ denotes the LD parameter, $\Gamma = \frac{2\pi}{\lambda}\Delta n d$ the phase difference, $\Delta n = n_e - n_o$ the linear OB, $\lambda$ the light wavelength in vacuum, $d$ the specimen thickness, $f_1$ and $f_2$ the functions of optical anisotropy and systematic error parameters, and $m_z$, $m_y$ the absorption coefficients referred respectively for the principal $z$ and $y$ directions in crystal. We stress here that the explicit appearance of the $f_1$ and $f_2$ functions is not important in our case. Eq. (2) may be obtained with a standard calculation of optical transmittance in the HAUP limits [5]. Using a thermo-optical effect, one can reach any phase difference for the fixed wavelength and, knowing the thickness of specimen, one can also obtain the absolute increment of the phase $\Gamma = 2\pi$ .

The optical transmission of the PA system should be measured prior to insertion of specimen into the optical scheme. The obtained data are approximated with a parabola and the intensity minimum is then calculated ($\left(\frac{\partial J}{\partial \chi}\right)_\theta = 0$) for every fixed polarizer azimuth. The intensity minima would lie on a straight line, with 45° slope angle in the ($\theta,\chi$) coordinate system. Then the same procedure should be performed for the PSA system. In this case the intensity minima also form a straight line, with the corresponding slope being equal to $\cos\Gamma/e^E$. The equation of this straight line in the ($\theta,\chi$) coordinate system is expressed as follows:

$$\chi(\theta)=\frac{\cos\Gamma}{e^{E}}\theta-\frac{1}{e^{E}}f_2 \quad . \qquad (3)$$

This slope angle is equal simply to $\cos\Gamma$ for non-dichroic crystals [9].

As a result, the slope of the straight line for the intensity minima should be measured for every specimen temperature. Eq. (3) is the first of principal definitions used within our method. However, in order to find both $\Gamma$ and $E$ values, it is necessary to know more than one equation. This is why the additional measuring procedures are used.

## 3. Analysis of HAUP maps

In a general case, Eq. (2) describes a second-order surface. Projections of the cuts of $J(\theta, \chi)$ surface by the planes $J =$ const have in general a shape of ellipses. The orientation of the latter in the $(\theta,\chi)$ coordinate system depends on the optical anisotropy parameters. A full set of these projections forms a HAUP map. Let us notice that in frame of the HAUP procedures, the similar map is represented in the coordinates $(\theta,Y)$, where $Y=\chi-\theta$ [4]. The angular slope of the principal axes of the ellipses on the HAUP map is equal to 45° for non-dichroic crystals, irrespective of the specimen temperature [9]. Besides, the eccentricity value changes with changing temperature. In case of dichroic crystals, the slope angle $\xi$ for the principal axes of the ellipses depends on temperature. In the $(\theta,\chi)$ coordinate system we have

$$\tan 2\xi=\frac{2\cos\Gamma}{e^{E}-e^{-E}} \, . \qquad (4)$$

Thus, after a completed process of polarizer and analyzer scanning, we analyze a 20×20 array of intensities collected for each temperature and all of the $E$ and Γ quantities. Then the corresponding second-order surface coefficients are obtained with the regression method. This makes it possible to find the unknown $E$ end Γ parameters. The optical anisotropy parameters are derived from the system of Eqs. (3) and (4). They are expressed as follows:

$$\begin{cases} E=\dfrac{1}{2}\ln\left(\dfrac{\tan 2\xi}{\tan 2\xi-2\partial\chi/\partial\theta}\right) \\ \cos\Gamma=\dfrac{\partial\chi}{\partial\theta}\sqrt{\dfrac{\tan 2\xi}{\tan 2\xi-2\partial\chi/\partial\theta}} \end{cases} . \qquad (5)$$

## 4. Results and discussion

The crystal plates under test were cut parallel to the optic axis and had the thickness of 0.72 mm. The HAUP maps and the $\chi(\theta)$ dependence were measured for the two positions of specimen in the PSA system, which were obtained with rotating the crystal around the beam propagation direction by 90°. This rotation of the specimen ensures a change in the sign of $E$ and Γ values.

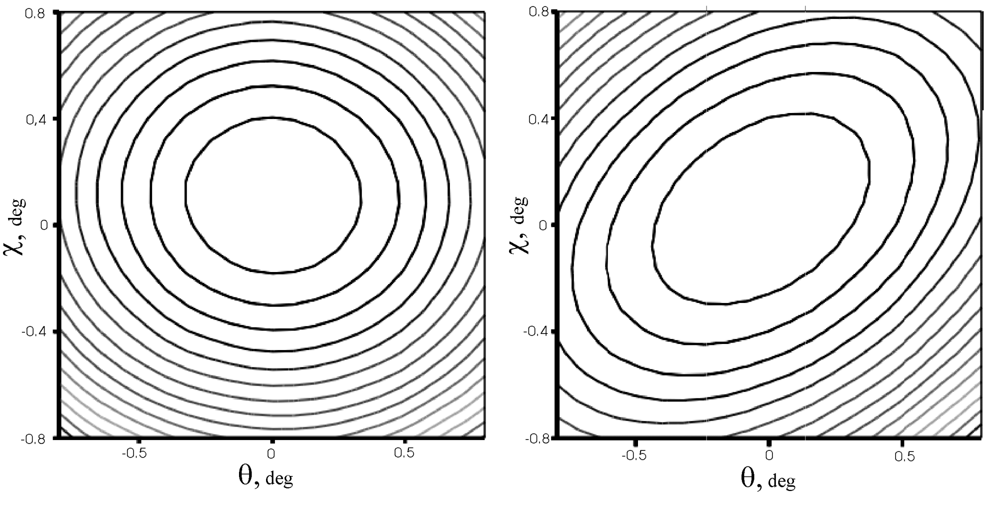

**Fig. 1.** Two examples of HAUP maps for the Cr-doped $Ca_3Ga_2Ge_4O_{14}$ crystals at different temperatures.

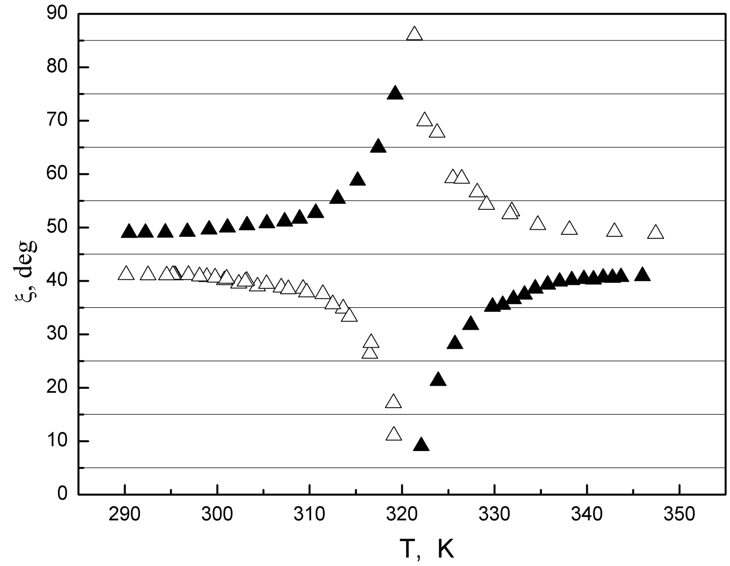

**Fig. 2.** Temperature dependence of angular slope $\xi$ for the principal axes of the ellipses observed on the HAUP maps for the two specimen positions (Δ, ▲) in the PSA system.

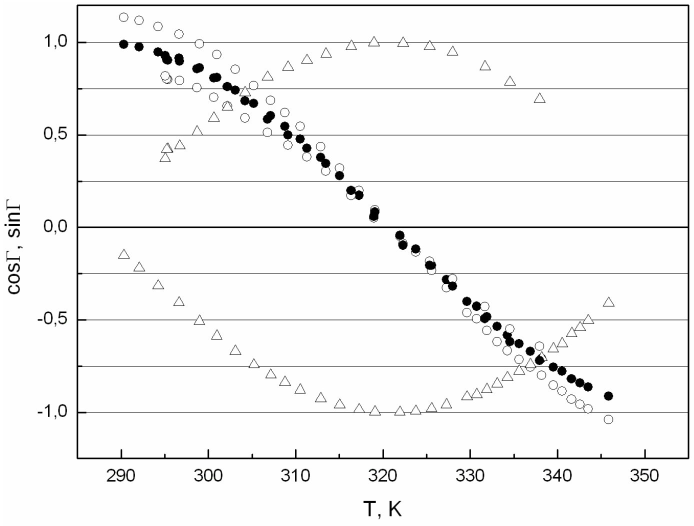

**Fig. 3.** Temperature dependences of $\cos\Gamma/e^{E}$ (○), $\cos\Gamma$ (●) and $\sin\Gamma$ (Δ) parameters for the two specimen positions in the PSA system.

This technique also enabled one to find the unknown systematic errors [9], though the latter do not influence the quantities $E$ and $\Gamma$, which are of interest in our case.

The HAUP maps in the $(\theta,\chi)$ coordinates derived for two different temperatures of crystal are illustrated in Fig. 1. The ellipses on these maps represent contour curves (the equi-intensify ellipses) of the measured light intensity. It is easy to see that the slope angles of principal axes of the ellipses differ from 45°. Temperature evolution of these slope angles for the two specimen positions in the PSA system is represented in Fig. 2.

Using a special computer program, we have obtained the temperature dependences of $\cos\Gamma$, the LD parameter $E$ and the OB changes $\delta(\Delta n)$ (see Fig. 3 and Fig. 4, respectively) on the basis of the straight $\chi(\theta)$ line parameters for the intensity minimum at each temperature and the calculated slope angles of principal axes of the ellipses. Some oscillations of the LD parameter are clearly seen in Fig. 4. They can be explained by influence of multiple beam reflections in our experiment, the effect described also in the works [10, 11]. In general, the problem of quantitative description of this effect is rather intricate. It is important at least that our experimental method and apparatus evidence such the effects. In this respect, it can really compete with the traditional techniques for measuring the LD.

The parameter $E$ obtained for our crystals is approximately equal to 0.13, while the difference between the absorption coefficients

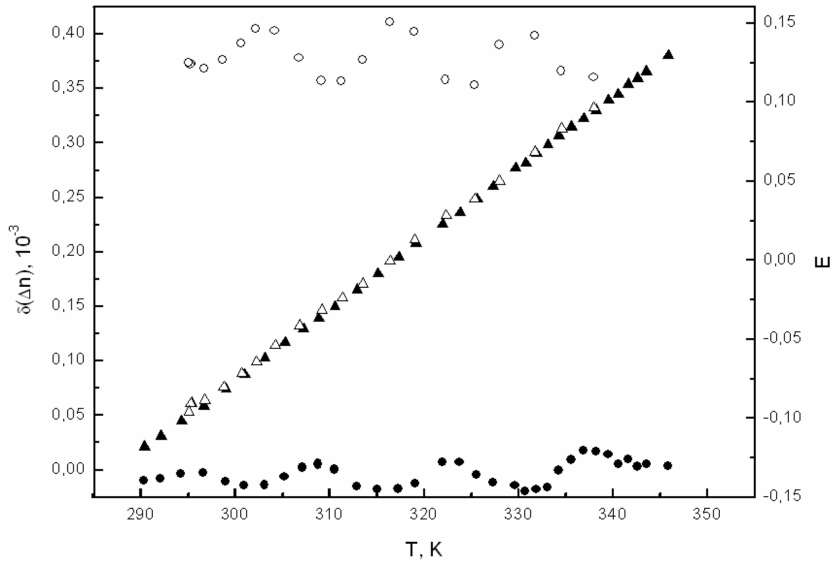

**Fig. 4.** Temperature dependences of LD parameter *E* (○,●) and OB changes (Δ, ▲) for the two specimen positions in the PSA system.

($\Delta m = m_z - m_y = 0.018 \cdot 10^{-3}$) is $10^3$ times less than the corresponding parameter derived in the work [5] for the crystals with considerable dichroism. Passing to a more frequently used coefficient $\Delta\kappa = 2\pi(m_z - m_y)/\lambda$, we get the value 1.80 cm$^{-1}$. Unfortunately, the exact concentration of Cr$^{3+}$ is not known in our case, thus hindering from further quantitative estimations of the effect of those ions.

Hence, we have successfully solved the experimental problem of crystal optics mentioned above, using a minimum amount of the measuring procedures. Employing additionally the information about the explicit form of $f_1$ and $f_2$ functions and the experimental results for the three characteristic azimuths $\theta_i$ ($i = 0$, 1 and 2), we are in a position to ascertain also the optical activity and the systematic error parameters.

Our specific single-wavelength experimental measurements of the LD have become possible owing to appreciable dichroic effect in Cr$^{3+}$-doped $Ca_3Ga_2Ge_4O_{14}$ crystals occurring just at $\lambda = 632.8$ nm. However, there are no fundamental limitations to its application in frame of more universal spectro-polarimetric techniques.

## Conclusions

1. The polarimetric method is suggested for simultaneous measurements of the OB and the LD, which is based on the HAUP-like maps and analyzing the slope parameters of the intensity minima data. The method is successfully applied to Cr$^{3+}$-doped $Ca_3Ga_2Ge_4O_{14}$ crystal.

2. The sensitivity of the measuring technique is enough for competing with the others methods known from the literature. In particular, the influence of multiple beam reflections on the measured value of LD parameter is observed.

3. The temperature dependences of the OB changes and the LD are obtained for the temperature region from 295 to 353 K.

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