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The Pockels effect in $TmAl_3(BO_3)_4$

V. A. Bedarev[a], D. N. Merenkov[a], M. I. Pashchenko[a], S. L. Gnatchenko[a], I. A. Gudim[b], L. N. Bezmaternykh[b], and V. L. Temerov[b]

[a]B. Verkin Institute for Low Temperature Physics and Engineering, National Academy of Sciences of Ukraine, Kharkov, Ukraine; [b]L.V. Kirensky Institute of Physics, Siberian Branch of Russian Academy of Sciences, Krasnoyarsk, Russian Federation

ABSTRACT

The Pockels effect was studied in $TmAl_3(BO_3)_4$ crystal. The electro-optic coefficient of the compound was determined. The principal refractive indexes of the crystal were determined by measuring the Brewster angles. Both contributions to the electric field induced birefringence associated with the redistribution of connected charges and deformation were estimated.

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Electric field; Pockels effect; birefringence; refractive indexes

Introduction

The noncentrosymmetric rare-earth aluminum borates $RAl_3(BO_3)_4$ demonstrate the magnetoelectric and piezoelectric properties[1–2]. For example, the magnetic field of 70 kOe induced the electric polarization $P = 300\ \mu C/m^2$ in $TmAl_3(BO_3)_4$, and giant $P = 3600\ \mu C/m^2$ in $HoAl_3(BO_3)_4$ [1–2]. Recently it was found that in the $TmAl_3(BO_3)_4$ crystal the electric field induces the linear birefringence which is directly proportional to the field (Pockels effect) [3].

It is known that optical indicatrix of $TmAl_3(BO_3)_4$ crystal with a point group 32 is an ellipsoid of rotation around the *c*-axis (C_3) with the principal refractive indexes n_a and n_c, measured along the *a*-axis (C_2) and *c*-axis respectively. The electric field $\boldsymbol{E}$ can reduce the optical class of crystal from the uniaxial to biaxial. In this case the symmetric part of the dielectric impermeability a_{ij} will have additives Δa_{ij}:

$$\Delta a_{ij} = r_{ijk}E_k + p_{ijlm}U_{lm}, \tag{1}$$

where E_k is the projection of the electric field vector on the coordinate axes. The tensor of electro-optic coefficients r_{ijk} is symmetric with respect to the *ij* indexes and describes the primary Pockels effect. This effect corresponds to the linear birefringence Δn_{el} caused by the redistribution of connected charges within the crystal. U_{lm} is a deformation of crystal and the photoelastic coefficient tensor p_{ijlm} is symmetric with respect to the *ij* and *lm* index pairs and describes the secondary Pockels effect caused by the electric field induced deformation of the piezoelectric crystal.

CONTACT V. A. Bedarev bedarev@ilt.kharkov.ua
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Let's consider the case when the electric field is directed along the a-axis. In the coordinate system $z||c$ and $x||a$ the matrix of coefficient r_{ijk} has the form:

$$r_{ijk} = \begin{vmatrix} r_{xxx} & 0 & 0 \\ r_{yyx} & 0 & 0 \\ 0 & 0 & 0 \\ r_{yzx} & 0 & 0 \\ 0 & r_{xzy} & 0 \\ 0 & r_{xyy} & 0 \end{vmatrix} \tag{2}$$

According to (2), the additives $r_{xxx}E_x$ and $r_{yyx}E_x$ to a tensor Δa_{ij} appear in an electric field $\boldsymbol{E}||a$. Considering that $r_{xxx} = -r_{yyx} = r$, the dependence of optical birefringence Δn_{el} on electric field $\boldsymbol{E}$ can be expressed as follows:

$$\Delta n_{el} = n_x - n_y = rn_a^3 E_x \tag{3}$$

In the same coordinate system the matrix of coefficient p_{ijlm} has the form:

$$p_{ijlm} = \begin{vmatrix} p_{xxxx} & p_{xxyy} & p_{xxzz} & p_{xxyz} & 0 & 0 \\ p_{yyyy} & p_{yyyy} & p_{yyzz} & p_{yyyz} & 0 & 0 \\ p_{zzxx} & p_{zzyy} & p_{zzzz} & 0 & 0 & 0 \\ p_{yzxx} & p_{yzyy} & 0 & p_{yzyz} & 0 & 0 \\ 0 & 0 & 0 & 0 & p_{xxzz} & p_{xzxy} \\ 0 & 0 & 0 & 0 & p_{xyxz} & p_{xyxy} \end{vmatrix} \tag{4}$$

In electric field the components of deformation U_{lm} related to the tensor of piezoelectric coefficients d_{lmk} as follow:

$$U_{lm} = d_{lmk}E_k \tag{5}$$

The form of piezoelectric tensor coefficients d_{lmk} is the same as the form of the tensor r_{ijk}. Application of an electric field $\boldsymbol{E}||a$ to the crystal causes a deformation with components: $U_{xx} = d_{xxx}E_x$; $U_{yy} = d_{yyx}E_x$. If $d_{xxx} = -d_{yyx}$, then $U_{xx} = -U_{yy}$. In response to (1) and (4), the linear birefringence caused by the deformation of the crystal in an electric field can be expressed as follows:

$$\Delta n_{def} = n_x - n_y = n_a^3(p_1 - p_2)U = n_a^3(p_1 - p_2)dE_x \tag{6}$$

where $p_1 = p_{xxxx} = p_{yyyy}$, $p_2 = p_{yyxx} = p_{xxyy}$, $U = U_{xx} = -U_{yy}$, $d = d_{xxx} = -d_{yyx}$.

As it was shown in the expressions (3) and (6), the general birefringence induced by the electric field in $TmAl_3(BO_3)_4$ crystal can be expressed as follows:

$$\Delta n = n_a^3(r + (p_1 - p_2)d)E_x = n_a^3(r + r_d)E_x = n_a^3 r_g E_x \tag{7}$$

where $r_d = (p_1\text{-}p_2)d$ – the electro-optical coefficient corresponding to the secondary electro-optical effect, and $r_g = (r + r_d)$ – the general electro-optical coefficient, depending both on primary and on secondary electro-optical effects.

For the correct establishment of electro-optical coefficients of $TmAl_3(BO_3)_4$ it is necessary to find the value of main refractive indexes. Furthermore, the measuring of two main refractive indexes allow to estimate contributions to the Pockels effect associated with the redistribution of connected charges and deformation.

The purpose of the present work is to establish the value of the electro-optical coefficient of the crystal $TmAl_3(BO_3)_4$, to determine the main refractive indexes of this crystal and to estimate the contribution of different mechanisms in the Pockels effect.

Experimental methods

For investigation of electro-optical effect the single crystal $TmAl_3(BO_3)_4$ sample was cut in the form of plane-parallel plate perpendicular to the *c*–axis. Typical single crystal plate dimensions are 4mm×4mm and thickness $t = 230\ \mu m$. The experiments were performed at room temperature.

The linear birefringence in electric field is related to the phase shift δ as follows:

$$\Delta n_E = \delta\lambda\ /\ 2\pi t \tag{8}$$

where λ is the light wavelength, t is thickness of sample. In the experiments a quarterwave plate was used as the compensator to measure the birefringence. The diagram of the experimental setup for δ-measurements based on Senarmont method described in [3]. The static electric field was created by the plates of the capacitor and the sample was placed inside of it. The direction of $\boldsymbol{E}$ coincides with the *a*–axis of crystal.

The angular dependences of the intensity of light reflected from the sample surface were measured to determine the main refractive indices of the crystal $TmAl_3(BO_3)_4$ at Brewster's geometry. It means that the polarization was in the incidence-reflection plane. Another sample was used for these studies. One of the sample planes was parallel to the *c*-axis and another one was perpendicular to this axis. When the light is reflected from a plane parallel to the *c*-axis, the Brewster angle φ_{Ba} was measured in the plane perpendicular to the *c*-axis connected with the main refractive index n_a by the simple relation:

$$n_a = \tan\varphi_{Ba} \tag{9}$$

For the light is reflected from a plane which is perpendicular to the c-axis, the Brewster angle φ_{Bc} is determined both n_a and n_c:

$$\tan\varphi_{Bc} = n_a\sqrt{\frac{n_c^2 - 1}{n_a^2 - 1}} \tag{10}$$

Results and discussion

The electric field dependence of birefringence in $TmAl_3(BO_3)_4$ is shown at Fig. 1. The figure shows that the sign of Δn_E depends on the electric field direction. The hysteresis was not found in experiments. The observed spontaneous optical activity is quite small ($\approx 0.3°$) and it does not influence the electro-optical effect [3].

The value of Δn_E reaches $8{\cdot}10^{-6}$ in an electric field $E = 4{\cdot}10^5$ V/m. Presented dependence is well extrapolated by the linear law:

$$\Delta n_E = \alpha E_x, \tag{11}$$

where $\alpha = 2{\cdot}10^{-11}$ m/V.

The angular dependences of the intensity of light reflected from two different planes of $TmAl_3(BO_3)_4$ crystal (Fig 2a, 2b) are well described by cubic function. The minima of these parabolas allow to determine the Brewster angles $\varphi_{Ba} = 55.3° \pm 0.15°$ and $\varphi_{Bc} = 58.1° \pm 0.15°$.

Substituting Brewster angles in equation (9) and (10) it is easy to obtain the values of the main refractive indexes of $TmAl_3(BO_3)_4$ crystal: $n_a = 1.44 \pm 0.01$ and $n_c = 1.53 \pm 0.01$. It makes possible to calculate the value of electro-optic coefficient $r_g = \alpha / n_a^3 \approx 6.6{\cdot}10^{-12}$m/V.

To compare the contributions of primary and secondary Pockels effect in electric-field-induced linear birefringence the expression (6) should be transformed as:

$$\Delta n_{def} = n_x - n_y = A'\frac{\Delta l}{l} \tag{12}$$

where $A' = n_a^3(p_1\text{-}p_2)$. It takes into account that U can be represented as relative deformation $\Delta l/l$ along the a-axis of crystal. Using (7) and (12) the electro-optical coefficient r_d can be expressed as:

$$r_d = \frac{A'd}{n_a^3} \tag{13}$$

The parameter d can be obtained by using the linear dependence of polariazation P along the a-axis of $TmAl_3(BO_3)_4$ from deformation $\Delta l/l$ [1]:

$$\frac{\Delta l}{l} = \frac{d}{\varepsilon_0(\varepsilon_x - 1)}P, \tag{14}$$

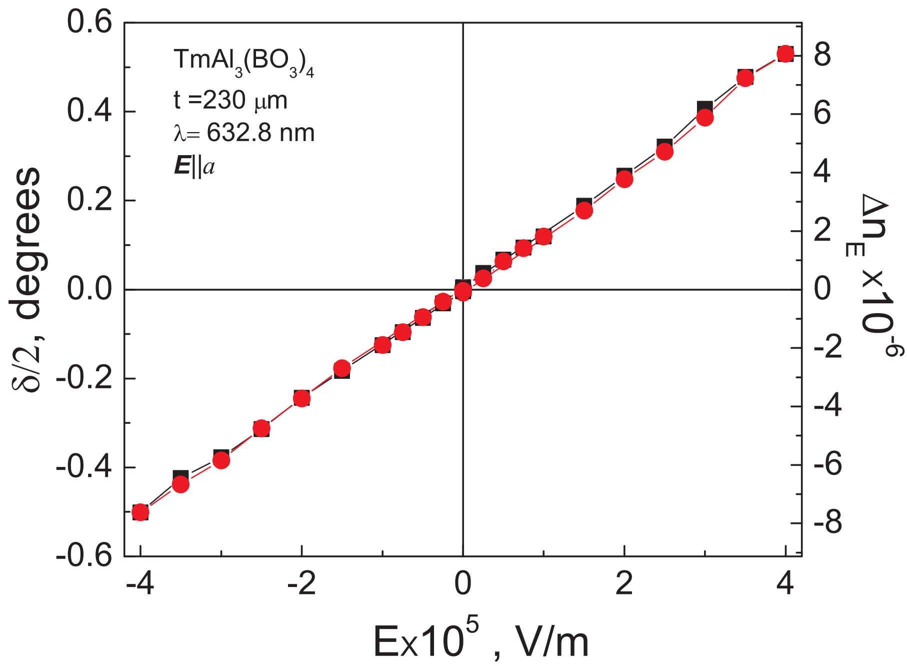

Figure 1. The linear birefringence of light as a function of the applied electric field in $TmAl_3(BO_3)_4$ at room temperature for $\boldsymbol{E}\|a$. The light beam with $\lambda = 632.8$ nm propagates along the c axis of the crystal. The thickness of the investigated sample is $t = 230$ μm.

here ε_x – the component of dielectric permittivity tensor of the crystal, ε_0 – vacuum permittivity. The measuring of the dielectric permittivity ε_x of $TmAl_3(BO_3)_4$ was carried out. The value of the measured $\varepsilon_x \approx 13.5$ is in a good agreement with the same value in samarium iron borate [5]. Taking into a count the experimental dates the parameter d was estimated from (14) as $d \approx 2.77{\cdot}10^{-11}$ m/V

The parameter A' in expression (13) is unknown but it can be estimated from the following considerations. The crystal structure of $TmAl_3(BO_3)_4$ can be considered in rhombohedral coordinate system. The axes of this system are on the edges of the elementary rhombohedron. Rhombohedron can be represented as the cube deformed along the C_3 axis. The length of the edges of the cube and the rhombohedron are the same, but the length of the diagonal parallel to the C_3 - axis of the cube and the rhombohedron are different. The length of the diagonal of a cube $C_{cub} = a\sqrt{3/2}$, where a the length of the edges of the cube. Then the relative difference of the lengths of the diagonals of the rhombohedron C_{romb} and cube C_{cub} can be determined by the following expression:

$$\Delta C = \frac{C_{cub} - C_{romb}}{C_{cub}} = \frac{a\sqrt{3/2} - C_{romb}}{a\sqrt{3/2}} \tag{15}$$

The optical properties of crystals with cubic and rhombohedral symmetry are different. For cubic symmetry the optical indicatrix of crystal is a sphere. For rhombohedral symmetry the optical indicatrix is an ellipsoid of revolution around the c-axis. And when the light

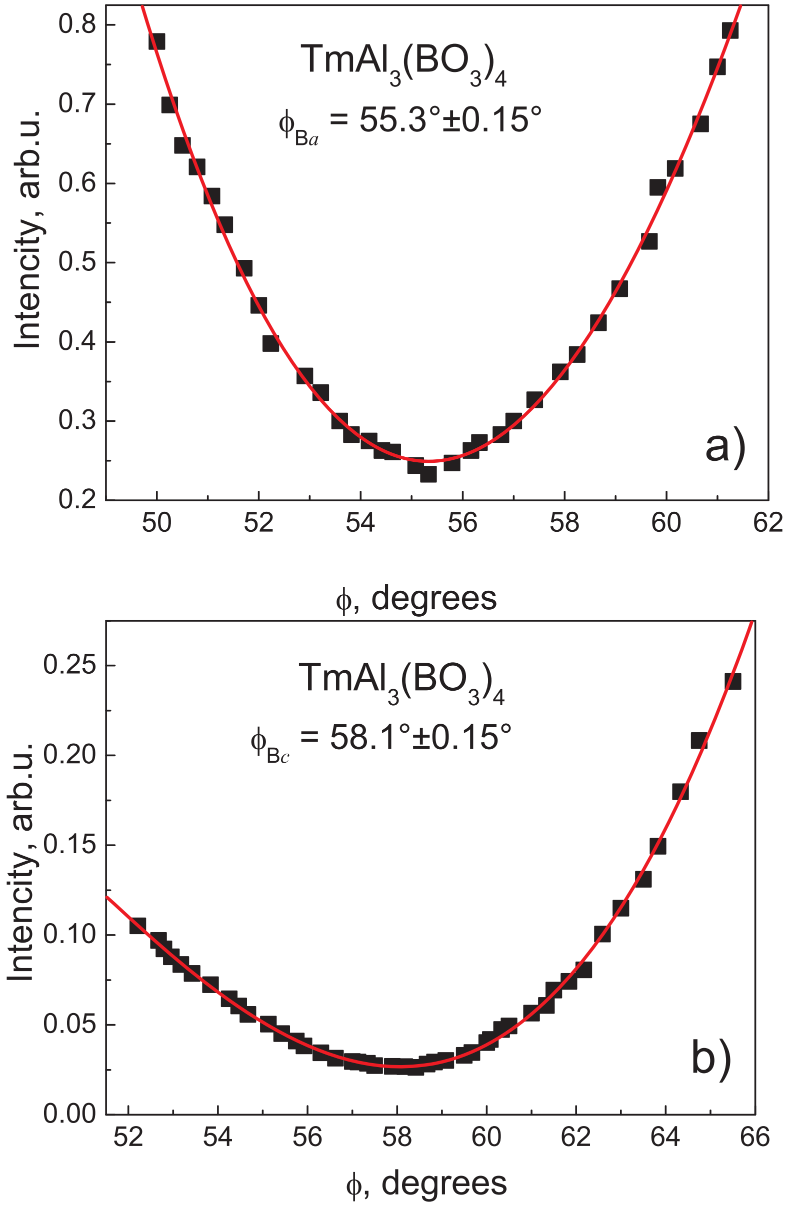

Figure 2. The intensity of the light reflected by the crystal planes of $TmAl_3(BO_3)_4$, which are parallel (a) and perpendicular (b) to the *c*-axis. φ the angle between the normal to the plane of crystal and incident light. The squares show the experimental data and the solid line approximation by cubic function.

propagates perpendicular to this axis the birefringence of light Δn_{cr} is observed in a crystal. The value of $\Delta n_{cr} = n_c - n_a$ depends on the parameter ΔC:

$$\Delta n_{cr} = A \cdot \Delta C = A \cdot \frac{a\sqrt{3/2} - C_{romb}}{a\sqrt{3/2}} \tag{16}$$

Here A is a proportionality coefficient. Using $C_{romb} = c = 7.218$ Å, $a = 9.274$ Å [4] and $\Delta n_{cr} = 0.09 \pm 0.01$ in (16) it's easy to obtain coefficient $A = 0.24 \pm 0.03$.

Assuming that $A \approx A'$ and using the corresponding parameters in expression (13) the value of coefficient of secondary Pockels effect was obtained $r_d \approx 2.2{\cdot}10^{-12}$ m/V. Thus, the value of coefficient of primary Pockels effect is $r \approx 4.4{\cdot}10^{-12}$ m/V. According to such estimation the contributions of primary and secondary Pockels effect to the electric field induced birefringence in $TmAl_3(BO_3)_4$ are within the same order of magnitude. But the primary Pockels effect is predominant in this crystal.

Conclusions

It was shown that the electric field induces linear birefringence in $TmAl_3(BO_3)_4$ crystal. The value of this electro-optical effect is proportional to the electric field. By measuring the angular dependences of the intensity of light reflected from the sample surface the Brewster angles $\varphi_{Ba} = 55.3° \pm 0.15°$ and $\varphi_{Bc} = 58.1° \pm 0.15°$ were determined and the values of the main refractive indexes of $TmAl_3(BO_3)_4$ crystal $n_a = 1.44 \pm 0.01$ and $n_c = 1.53 \pm 0.01$ were obtained. The general electro-optic coefficient of $TmAl_3(BO_3)_4$ crystal $r_g \approx 6.6{\cdot}10^{-12}$ m/V was established. And the primary and secondary Pockels effects coefficients were estimated as $r \approx 4.4{\cdot}10^{-12}$ m/V and $r_d \approx 2.2{\cdot}10^{-12}$ m/V respectively.

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