About the Possibility of Linearization of Quadratic Cotton-Mouton Birefringence Change due to *kH*-Effect. The Case of CdS Crystals

Vlokh R., Adamenko D., Klymiv I. and Vlokh O.

Institute of Physical Optics, 23 Dragomanov Str., 79005, Lviv, Ukraine
e-mail: vlokh@ifo.lviv.ua

Received: 22.03.2004

Abstract

A possibility for appearance of the so-called *kH*-effect in crystals is analyzed, which originates from the light absorption, spatial dispersion and the influence of magnetic field. It is shown that the *kH*–effect can lead to linearization of the Cotton-Mouton effect. The coefficients of both the Cotton-Mouton ($(n_3^3\beta_{31} - n_1^3\beta_{11}) = 2.08\times10^{-15} Oe^{-2}$) and the *kH*-effect ($\delta_{1112} \le 0.72\times10^{-20} m/Oe$) are obtained for CdS crystals.

Key words: nonreciprocal birefringence, spatial dispersion, magnetooptics

PACS: 78.20.Ls

Introduction

In our previous report [1] we have shown that accounting for the optical energy dissipation and the spatial dispersion leads to a possibility for new parametric crystal optical effects induced with magnetic field in crystals not manifesting magnetic ordering. Under the presence of external magnetic field, spatial dispersion and the absorption, optical-frequency dielectric constant may be written as

$$\varepsilon_{ij}=\varepsilon_{ij}^{(o)}+i\varepsilon_{ij}`=\varepsilon_{ij}+i\varepsilon_{ij}`+i\alpha_{ijm}H_m-\alpha_{ijm}`H_m+i\gamma_{ijk}k_k-\gamma`_{ijk}k_k-\boldsymbol{\delta_{ijkm}k_kH_m}-\boldsymbol{i\delta`_{ijkm}k_kH_m}. \quad (1)$$

Here the first two terms ε_{ij} and $i\varepsilon_{ij}`$ describe the ordinary refraction and the absorption; the third ($i\alpha_{ijm}H_m$) and the fourth ($\alpha_{ijm}`H_m$) ones the Faraday effect and the refractive index change proportional to the magnetic field in dissipative media, respectively, the $i\gamma_{ijk}k_k$ and $\gamma`_{ijk}k_k$ terms refer respectively to the gyration and non-reciprocity of the refractive indices in dissipative gyrotropic media, the term $\boldsymbol{\delta_{ijkm}k_kH_m}$ corresponds to non-reciprocity in the refractive indices in gyrotropic dissipative media in the presence of magnetic field, and, finally, the term $\boldsymbol{-i\delta`_{ijkm}k_kH_m}$ produces a magnetogyration effect. The magnetogyration effect has been therefore considered as an addition to the effect of non-reciprocity of the refractive indices in gyrotropic media inside a spectral range of essential absorption under the application of magnetic field. The coefficient $\delta`_{ijkm}$ is a fourth-rank axial (or pseudo-) tensor antisymmetric in *i* and *j* indices, which may be rewritten according to the duality principle as $\delta`_{ijkm}=e_{ijr}\eta_{rkm}$, where e_{ijr} stands for the Levi-Civit's unit pseudo-tensor and η_{rkm} a third-rank polar tensor with the internal symmetry V^3. It has been experimentally proved in the studies [1,2] that the magnetogyration effect, discovered more than twenty years ago [3-8], exists only in crystals with considerable absorption. On the other side, as reported by *Krichevtsov, Pisarev et. al.* [9], an effect that follows directly from Eq. (1) has been observed in $Cd_{1-x}Mn_xTe$ crystals (the point group of symmetry $\bar{4}3m$): the so-called *kH*-effect ($\Delta n \propto \delta_{ijkm}k_kH_m$) being additional to the well-known Cotton-Mouton change in the refractive indices. The authors recur to a quite unusual interpretation of the observed phenomena even on phenomenological level. Let us consider the relation suggested in [9] for the description of those phenomena:

$$\Delta\varepsilon_{ij} = \gamma^s_{ijkl}B_kk_l + g_{ijs}[B \times k]_s \qquad (2)$$

where $\Delta\varepsilon_{ij}$ represents the change of the dielectric permittivity, B_k the magnetic displacement, k the wave vector of light, γ^s_{ijkl} the part of fourth-rank axial tensor symmetric under interchange of the first and last pairs of indices (the latter statement is according to [9]), and g_{ijs} the fully symmetric third-rank polar tensor (also according to [9]).

First of all, the tensor γ^s_{ijkl} is symmetric only under the permutation of the first two indices, because the indices k and l refer to different physical quantities (the magnetic field and the wave vector, respectively). Secondly, the tensor g_{ijs} is not fully symmetric, being in the best case symmetric only in the first two indices. Moreover, it is impossible to find the *kH*-effect (the "*Bk*-effect", in the notations by the authors of [9]) in the experimental geometry used in [9] (k || [110] and B || [001]), since all the relevant components of γ^s_{ijkl} tensor are equal to zero and the scalar products B_3k_1 and B_3k_2 do not give rise to any perturbation of optical indicatrix. As for the vector products $[B_3k_1]$ and $[B_3k_2]$, unfortunately, the authors [9] have not explained a physical meaning of polar vector that appears through vectorial multiplication of the polar vector k and the axial vector B. Obviously, that should be some polar vector M_s, though its physical implications are still not clear. Moreover, taking the polar vectors $M_{1,2}=[B_3k_{2,1}]$ as external field quantities contradicts principled symmetry laws (namely, the *Curie* and the *Neumann* principles), while leading to crystal symmetry breaking to the triclinic point group 1. Lowering crystal symmetry occurs only under the magnetic field action (the limiting symmetry group ∞/m). Then we have $\bar{4}3m \bigcap \infty_3/m = 2$, i.e. a lowering of the point group to the monoclinic one 2 (2 || [001]). Even if we suppose that the polar vectors $M_{1,2}$ act and write the equation of optical indicatrix,

$$\begin{aligned}&(B_o + \beta_{12}H_3^2)x^2 + (B_o + \beta_{12}H_3^2)y^2 + \\ &+(B_o + \beta_{11}H_3^2)z^2 + 2g_{41}M_1zy + \\ &+2g_{41}M_2zx = 1\end{aligned} \quad , \qquad (3)$$

where B_o is the optical impermeability constants and β_{ij} the Cotton-Mouton tensor, one could easily see that the change in the principal refraction indices (as well as the induced birefringence) would be proportional to H^2 and $\left(M_1^2 + M_2^2\right)$ but by no means to M or H.

The aim of the present paper is to re-investigate the *kH*-effect ($\Delta n \propto \delta_{ijkm}k_kH_m$), which should appear as a phenomenon accompanying the well-known Cotton-Mouton refractive index change, on the example of CdS crystals.

Experimental Approach and Results

In order to study the induced birefringence, we have used the well-proven Senarmont method and high-quality homogeneous CdS crystals (the point symmetry group *6mm*). One can assume that the Cotton-Mouton effect might be large enough in comparison with the linear additional effect that appears owing to optical absorption and spatial dispersion. Thus, we should expect the appearance of a weak linearization of quadratic (Cotton-Mutton) dependence of the birefringence increment on the magnetic field.

Let us analyze the corresponding tensors and the optical indicatrix equation in the presence of three different effects – the Cotton-Mouton effect and the two linear magnetooptic effects governed with the $\Delta n \propto \alpha_{ijm}`H_m$ and $\Delta n \propto \delta_{ijkm}k_kH_m$ terms. The Cotton-Mouton effect is described by a polar fourth-rank tensor with the internal symmetry $[V^2]^2$, whose form for the point group *6mm* is as follows:

	H_1H_1	H_2H_2	H_3H_3	H_3H_2	H_3H_1	H_2H_1
ΔB_{11}	β_{11}	β_{12}	β_{13}	0	0	0
ΔB_{22}	β_{12}	β_{11}	β_{13}	0	0	0
ΔB_{33}	β_{31}	β_{31}	β_{33}	0	0	0
ΔB_{32}	0	0	0	β_{44}	0	0
ΔB_{31}	0	0	0	0	β_{44}	0
ΔB_{21}	0	0	0	0	0	β_{44}

.The linear magnetooptic effect may be understood in terms of a third-rank axial tensor possessing the symmetry $\varepsilon[V^2]V$:

	H_1	H_2	H_3
ΔB_{11}	0	0	0
ΔB_{22}	0	0	0
ΔB_{33}	0	0	0 ,
ΔB_{32}	α_{41}	0	0
ΔB_{31}	0	$-\alpha_{41}$	0
ΔB_{21}	0	0	0

and, finally, the bi-linear *kH*-effect is described by an axial fourth-rank tensor with the symmetry $\varepsilon[V^2]V^2$:

	k_1H_1	k_2H_2	k_3H_3	k_3H_2	k_3H_1	k_2H_1	k_2H_3	k_1H_3	k_1H_2
ΔB_{11}	0	0	0	0	0	δ_{1121}	0	0	δ_{1112}
ΔB_{22}	0	0	0	0	0	$-\delta_{1121}$	0	0	$-\delta_{1112}$
ΔB_{33}	0	0	0	0	0	0	0	0	0
ΔB_{32}	0	0	0	0	δ_{3231}	0	0	δ_{3213}	0
ΔB_{31}	0	0	0	$-\delta_{3231}$	0	0	$-\delta_{3213}$	0	0
Δa_{21}	δ_{1111}	δ_{1122}	0	0	0	0	0	0	0

It is seen from these tensors that the Cotton-Mouton effect and the *kH*-effect would lead to changes in the principal refractive indices (without any rotations of the optical indicatrix) when the magnetic field is applied along *Y* axis in CdS crystals and the light propagates along *X* axis, while the optical indicatrix would rotate around the *Y* axis due to the linear magnetooptic effect. The corresponding optical indicatrix equation results in

$$\begin{aligned}&(B_1+\beta_{12}H_2^2+\delta_{1112}k_1H_2)X^2+\\&+(B_1+\beta_{11}H_2^2-\delta_{1112}k_1H_2)Y^2+\\&(B_3+\beta_{31}H_2^2)Z^2-2\alpha_{41}H_2ZX=1\end{aligned}\quad. \tag{4}$$

After rewriting Eq. (1) in the proper frame of reference we obtain

$$\begin{aligned}&(B_1+\beta_{12}H_2^2+\delta_{1112}k_1H_2+\frac{\alpha_{41}^2H_2^2}{\left[(B_1-B_3+\beta_{12}H_2^2-\beta_{31}H_2^2)+\delta_{1112}k_1H_2\right]})X^2+\\&(B_1+\beta_{11}H_2^2-\delta_{1112}k_1H_2)Y^2+\\&(B_3+\beta_{31}H_2^2-\frac{\alpha_{41}^2H_2^2}{\left[(B_1-B_3+\beta_{12}H_2^2-\beta_{31}H_2^2)+\delta_{1112}k_1H_2\right]})Z^2=1\end{aligned}\quad. \tag{5}$$

The expressions for the principal refractive indices may be represented as

$$\begin{aligned}&n_1'=n_1-\frac{1}{2}n_1^3\left\{\beta_{12}H_2^2+\delta_{1112}k_1H_2+\frac{\alpha_{41}^2H_2^2}{\left[(B_1-B_3+\beta_{12}H_2^2-\beta_{31}H_2^2)+\delta_{1112}k_1H_2\right]}\right\}\\&n_2'=n_1-\frac{1}{2}n_1^3\left\{\beta_{11}H_2^2-\delta_{1112}k_1H_2\right\}\\&n_3'=n_3-\frac{1}{2}n_3^3\left\{\beta_{31}H_2^2-\frac{\alpha_{41}^2H_2^2}{\left[(B_1-B_3+\beta_{12}H_2^2-\beta_{31}H_2^2)+\delta_{1112}k_1H_2\right]}\right\}\end{aligned}\quad. \tag{6}$$

If the optical beam is propagated along the X direction, the birefringence increment induced with the magnetic field H_2 becomes

$$\delta(\Delta n)_{23} = -\frac{1}{2}\left\{(n_1^3\beta_{11} - n_3^3\beta_{31})H_2^2 - n_1^3\delta_{1112}k_1H_2 + n_3^3\frac{\alpha_{41}^2H_2^2}{\left[(B_1 - B_3 + \beta_{12}H_2^2 - \beta_{31}H_2^2) + \delta_{1112}k_1H_2\right]}\right\} \approx \frac{1}{2}\left\{(n_3^3\beta_{31} - n_1^3\beta_{11})H_2^2 + n_1^3\delta_{1112}k_1H_2\right\}. \quad (7)$$

It is possible to neglect the smallest last term in the r.h.s. of Eq. (7) and so the resulting birefringence increment reduces to

$$\delta(\Delta n)_{23} = \frac{1}{2}(n_3^3\beta_{31} - n_1^3\beta_{11})H_2^2 + \frac{1}{2}n_1^3\delta_{1112}k_1H_2. \quad (8)$$

One can see that the sign of the additional *kH*-effect should depend on the direction of light propagation, i.e. the sign of the wave vector. Thus, contrary to the Cotton-Mouton effect, the *kH*-effect turns out to be *k*-sensitive. It is reasonable to remember that, in case of experimental separation of the Faraday effect and the *k*-sensitive magnetogyration [1,2], the operation of sign change for the wave vector ($k_z \to -k_z$) has been performed with the rotation of sample by 180° around the axis perpendicular to the Z axis. Such the operation leads to the change of sign of the third-rank polar magnetogyration tensor, whereas the sign of the polar second-rank Faraday tensor remains unaltered in the same coordinate system. One can try to apply the same operation for checking experimentally the fact whether the observable appearance of the linear birefringence in the magnetic field really corresponds to the *kH*-effect.

Let us analyze what would happen if CdS sample is rotated by 180° around the Z axis under the conditions that the light is propagated along X or X axes and the magnetic field is applied along the Y direction. It is easy to check that, contrary to the magnetogyration tensor, the tensor components of both the Cotton-Mouton $(n_3^3\beta_{31} - n_1^3\beta_{11}) \to (n_3^3\beta_{31} - n_1^3\beta_{11})$ and the

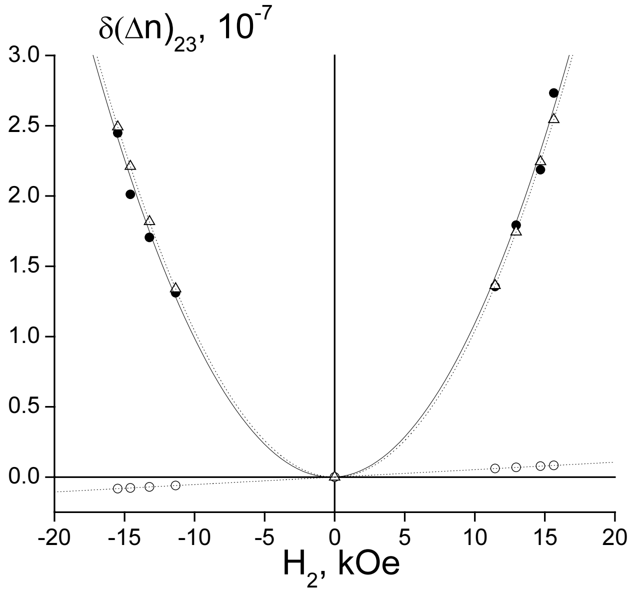

Fig. 1. Dependence of the birefringence increment versus magnetic field for CdS crystal at λ=632.8nm: solid curve (full circles) – experimental data; dashed curve (open triangles) – pure Cotton-Mouton effect; dashed line (open circles) – pure *kH*-effect.

kH–effects ($\delta_{1112} \to \delta_{1112}$) keep their signs invariable under this operation. It means that, after rotating the sample by 180^o around the *Z* axis, there will be no difference between the rotation directions of polarization plane φ_1 behind the quarter-wave plate, appearing due to Cotton-Mouton and *kH*-effects. Instead, a linearization of the Cotton-Mouton effect could be visible if the latter is really accomplished with the *kH*-effect.

As seen from Figure 1, the manifestation of the effects described above is fairly well confirmed experimentally. The obtained dependences have been fitted with the function $\delta(\Delta n)_{23} = A + BH + CH^2$, where *A*=–0.02366, *B*=0.0053 and *C*=0.0104.

The following coefficients of the Cotton-Mouton and *kH*-effects may be calculated from those dependences:

$$(n_3^3\beta_{31} - n_1^3\beta_{11}) = 2.08 \times 10^{-15}\, Oe^{-2},$$

$$\delta_{1112} \le 0.72 \times 10^{-20}\, m/Oe.$$

One can see that the *kH*-effect is very small. The value of the induced birefringence ($\sim 10^{-8}$) due to this effect is only slightly above our experimental sensitivity. It is therefore difficult to testify unambiguously whether the feeble linearization of the Cotton-Mouton effect observed by us is really the *kH*-effect.

Conclusion

Based on the analysis reported in this work, one can conclude that, from the viewpoint of electrodynamic equations, the light absorption, spatial dispersion and the effect of external magnetic field should produce the observable *kH*-effect. On the other hand, the experimental search of this effect itself is associated with the necessity of increasing the birefringence increment sensitivity up above 10^{-8}. It follows from our experiments that the values of the Cotton-Mouton and *kH*-effect coefficients for CdS crystals are respectively $(n_3^3\beta_{31} - n_1^3\beta_{11}) = 2.08 \times 10^{-15}\, Oe^{-2}$ and $\delta_{1112} \le 0.72 \times 10^{-20}\, m/Oe$ at λ=632.8nm .

Acknowledgement

We would like to acknowledge the financial support of this study from the Ministry of Education and Science of Ukraine under the Project N0103U000701.

Reference

1. Vlokh R., Vlokh O.G., Klymiv I., Adamenko D. Ukr. J. Phys. Opt. **3** (2002) 166.
2. Vlokh R., Vlokh O.G., Klymiv I., Adamenko D. Ukr. J. Phys. Opt. **3** (2002) 271.
3. Vlokh O.G. Bulletin Lviv Univ. **16** (1982) 5 (in Ukrainian).
4. Zheludev I.S., Vlokh O.G., Sergatyook V.A. Ferroel. **63** (1985) 97.
5. Vlokh O.G., Sergatyook V.A. Rep. USSR Acad. Sci. **291** (1986) 832.
6. Vlokh O.G., Sergatyook V.A. Ferroel. **80** (1988) 313.
7. Vlokh O.G., Vlokh R.O. Opt. Spektrosk. **69** (1989) 225.
8. Vlokh O.G., Vlokh R.O. Opt. Spektrosk. **69** (1990) 458.
9. Krichevtsov B.B., Pisarev R.V., Rzhevsky A.A. et. al. Phys. Rev. B **57** (1998) 14611.