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ON THE POSSIBILITY OF MEASUREMENTS OF NONLINEAR ELECTROOPTIC EFFECTS IN 432 SYMMETRY CRYSTALS

It is shown that despite the fact that in 432 point group crystals the linear electrooptic effect is vetoed by symmetry there are configurations for which the third-order effect may appear. The results obtained indicate that the 432 crystals are promising media for measurements of nonlinear electrooptic effects. Macroscopic description of nonlinear electrooptic phenomena in the crystals is presented and the matrices of tensors representing the effects up to the fifth-order are given.

Keywords: nonlinear electrooptic effects, matrices of electrooptic tensors, 432 symmetry crystals.

1. INTRODUCTION

Experimental results have been published for a variety of electrooptic crystals. Among them several results comprising fourth-order electrooptic phenomena have been presented. However, in respect of the nonlinear electrooptic effects a large spread is observed in the relevant coefficients of a given crystal (see, e.g. [1, 2]).

Theoretical description of the macroscopic conditions for measurements of the fourth-order electrooptic effect have been presented in Refs. [3, 4]. Moreover, some theoretical results related to the fifth-order effect have been reported [5]. Recently, some configurations allowing for measurements of the third-order (cubic) or higher-orders electrooptic phenomena have been indicated

[6, 7]. However, up to now, configurations allowing for measurements of the third-order effect in 432 symmetry crystals have not been considered.

The nonlinear susceptibilities responsible for electrooptic effects originate from two sources. The first one is the lattice contribution resulting from the interaction of the applied electric field with the crystal lattice, which in turn changes the electronic polarizability. The second one is a purely electronic term in which the electronic polarizability is directly altered by the applied field (see, e.g. [8]). The lattice contribution is responsible, for example, for the first- and second-order Raman scattering, hyper-Raman scattering and the optical rectification. The purely electronic term contributes, for example, to second- and third-harmonic generation, electric-field-induced first-order Raman scattering, the waves mixing and nonlinear refractive index. Investigations of the electrooptic phenomena complement the data obtained in research on nonlinear optics, electrical and optico-electrical phenomena. The fact that the electrooptic effects have the same origin as numerous physical phenomena covering the wide field of the solid state physics, nonlinear optics and spectroscopy is the reason that its experimental and theoretical investigations are of great interest.

Traditionally, in a field free principal axes system, electrooptic coefficients are defined as terms in a power-series expansion in the low-frequency electric field $\boldsymbol{E}$ of the optical frequency impermeability tensor $\eta_{ij}(\omega)$. The coefficients of the electrooptic effects are partial derivatives of the components of the impermeability tensor in relation to n components of the electrical field (see, e.g. [9-11]). The tensor $K_{ijk_1...k_f}$ that represents the electrooptic effect of the order f is given by

$$K_{ijk_1...k_f} = \frac{1}{f!}\left(\frac{\partial^f \eta_{ij}(\omega)}{\partial E_{k_1}...E_{k_f}}\right). \tag{1}$$

The tensor $K_{ijk_1...k_f}$ is symmetrical in relation to the first pair of indices (i,j) and the other indices (k_1, k_2, $\ldots k_f$). The first property is the consequence of the symmetry of the impermeability tensor; the second is due to the symmetry of multivector E_1, E_2, $\ldots E_f$. The tensor $K_{ijk_1...k_f}$ related to the f-th-order electrooptic effect constitutes the tensor of $n = f + 2$ rank with the internal symmetry $[V^2][V^f]$.

According to a fundamental postulate of crystal physics, known as Neumann`s Principle, the symmetry of any physical property of a crystal must include the symmetry elements of the point group of the crystal [12]. The effect of crystal symmetry on physical properties represented by tensors, results in changes in matrices of the tensors. Any element of a point symmetry group may reduce the number of independent and non-zero components. For example, no physical property that is represented by an odd rank tensor can appear in a centrosymmetric crystal. This means that all components of the tensor are equal to zero. The opposite statement, however: in a noncentrosymmetric crystal any property represented by an odd rank tensor may appear does not have to hold true. Nevertheless, the fact that the above statement is nearly always true leads sometimes to mistakes. Good examples of such a case are the converse piezoelectric and the linear electrooptic effects which are represented by the tensor of internal symmetry $[V^2]V$. Despite the fact that the 432 symmetry crystals lack the inversion symmetry, the other symmetry elements make all the components of the tensor $[V^2]V$ equal to zero.

The fact that in the 432 symmetry crystals the linear electrooptic effect is vetoed by the symmetry suggests that the crystals are promising media for attempts to measure higher-order electrooptic effects that hitherto have not been observed. This is of special interest as measurements of nonlinear electrooptic effects may be affected by a contribution of the linear response. It has been shown that even in configurations theoretically forbidding the linear contribution, unavoidable experimental disorientations, not perfect quality of crystals, or an angular divergence of light can lead to a situation when the nonlinear response is observed on a background of the linear one or the linear effect manifests as a nonlinear response [4, 13].

The aim of this work is to consider the existence of configurations which allow for measurements of the third-order electrooptic effect in the 432 symmetry class crystals.

2. TENSORS REPRESENTING ELECTROOPTIC EFFECTS OF VARIOUS ORDERS IN 432 SYMMETRY CRYSTALS

In this work the shortened, widely used, matrix notation is applied for the symmetrical tensor of the second rank [12], namely;

$$ij \rightarrow \mu,$$

where: $$11\rightarrow 1,\ 22\rightarrow 2,\ 33\rightarrow 3,\ 23\rightarrow 4,\ 13\rightarrow 5,\ 12\rightarrow 6 \qquad (2)$$

Thus the shortened matrix notations for the electrooptic tensors of the linear, quadratic, third-, fourth- and fifth-orders are, respectively,

$$r_{ijk} = r_{\mu k} ,$$

$$g_{ijkl} = g_{\mu\lambda} ,$$

$$R_{ijklm} = R_{\mu klm} \tag{3}$$

$$K_{ijklmn} = K_{\mu klmn} ,$$

$$L_{ijklmno} = L_{\mu klmno} .$$

To describe the electrooptic response of the 432 symmetry crystals the potential tensor functions have been used. With the use of common invariants of the symmetrical tensor of second rank and a vector obtained by Smith et al [14], the function describing the dependence of the impermeability tensor up to the fifth power of the electrical field for the 432 symmetry class was obtained. The enumeration of the pertinent partial derivatives produced relations between non-zero components.

The numbers of independent components of the tensors agree with those obtained by group theory [15]. The numbers were computed using the formula

$$n = \frac{1}{N}\sum_{g\epsilon F} \chi^0 (g), \tag{4}$$

where F is the point group of a crystal containing symmetry elements g, N is the order of the group F and the character of matrix transformation $\chi^0(g)$ is expressed by [16]

$$\chi^0(g) = \frac{1}{N_p}\sum_{p\epsilon P} \chi\left(g^{l_1}\right)\chi\left(g^{l_2}\right) \ldots \chi\left(g^{l_f}\right), \tag{5}$$

where P is the group comprising a set of all such permutations for which the internal symmetry is taken into account, N_p is the order of the group P, $\chi(g^l)$ is the character of transformation representing the element symmetry g in the vector representation of the group F, $l_1 \ldots l_f$ are the lengths of corresponding cycles of permutations and the summation extends over all elements in P.

The results obtained for the matrices of tensors related to the electrooptic phenomena up to the fifth-order are presented in Tables 1-5 showing indices of the nonzero components of the tensors. The matrices of tensors representing the linear and quadratic electrooptic effects agree of course with those widely known from handbooks (see, e.g. Refs [9-12]). Nevertheless, for the sake of clarity we remind them here. Moreover, the tensors representing the fourth-order electrooptic effect have been previously obtained for all symmetries [3]. These are reminded for the 432 crystals in Table 4.

Table 1

The form of the tensor $r_{\mu k}$ of the internal symmetry $[V^2][V]$ in the 432 symmetry class (see, e.g. Refs [9-12])

The number of non-zero and independent components	The components of the tensor
0, 0	All vanish

Table 2

The form of the tensor $g_{\mu\lambda}$ of the internal symmetry $[V^2][V^2]$ in the 432 symmetry class (see, e.g. Refs [9-12]), the shortened matrix notation given in Eqs (2) and (3) is used

The number of non-zero and independent components	The components of the tensor
12, 3	$11 = 22 = 33 = g_{11}$ $12 = 13 = 21 = 23 = 31 = 32 = g_{12}$ $44 = 55 = 66 = g_{44}$

Table 3

The form of the tensor $R_{\mu klm}$ of the internal symmetry $[V^2][V^3]$ in the 432 symmetry class, this work, the shortened matrix notation given in Eqs (2) and (3) is used

The number of non-zero and independent components	The components of the tensor
6, 1	$4133 = -4122 = 5112 =$ $= -5233 = 6223 = -6113 = R$

Table 4

The form of the tensor $K_{\mu klmn}$ of the internal symmetry $[V^2][V^4]$ in the 432 symmetry class, compare Ref. [3], the shortened matrix notation given in Eqs (2) and (3) is used

The number of non-zero and independent components	The components of the tensor
27, 6	$11111 = 22222 = 33333 = K_1$ $12233 = 21133 = 31122 = K_2$ $12222 = 23333 = 31111 =$ $=13333 = 21111 = 32222 = K_3$ $11133 = 11122 = 22233 =$ $= 21122 = 32233 = 31133 = K_4$ $42223 = 43332 = 51113 =$ $= 53331 = 61112 = 62221 = K_5$ $41123 = 52213 = 63312 = K_6$

Table 5

The form of the tensor $L_{\mu klmno}$ of the internal symmetry $[V^2][V^5]$ in the 432 symmetry class, this work, the shortened matrix notation given in Eqs (2) and (3) is used

The number of non-zero and independent components	The components of the tensor
18, 3	112223 = -112333 = 212333 = = -211123 = 311123 = -312223 = L_1 411122 = -411133 = 522233 = = -511112 = 611333 = -622333 = L_2 412222 = -413333 = 523333 = = -511112 = 611113 = -622223 = L_3

3. DISCUSSION

As an example we consider the following direction of the electric field: $\sqrt{2}\mathbf{E} = (E,0,E)$. The results presented in Tables 1-5 show that, considering the terms up to the fifth power of the electric field, the new refractive indices are:

$$1/n_1'^2 = 1/n_o^2 + (g_{11} + g_{12})E^2 + (K_1 + K_3 + 6K_4)E^4$$

$$1/n_2'^2 = 1/n_o^2 + 2g_{12}E^2 + (K_2 + 2K_3)E^4$$

$$1/n_3'^2 = 1/n_o^2 + (g_{11} + g_{12})E^2 + (K_1 + K_3 + 6K_4)E^4 \qquad (6)$$

$$1/n_4'^2 = 3RE^3 - 5(L_2 + L_3)E^5$$

$$1/n_5'^2 = 2g_{44}E^2 + 8K_5E^4$$

$$1/n_6'^2 = -3RE^3 + 5(L_2 + L_3)E^5,$$

where n_o is the field-free refractive index. With the exception of the factor 2 appearing twice in the second equation of the set labeled as Eq. (6), the other numerical factors result from the symmetry of the multivector E_1, E_2, …E_f. For example, $E_1E_3E_3 = E_3E_1E_3 = E_3E_3E_1$, thus in the fourth equation of the set the term 3R appears. Eq. (6) shows that for the considered direction of the electric field, the third-order electrooptic effect may be experimentally determined with the light beam propagating along the X_1 or X_3 directions. The third-order electrooptic effect is responsible for rotations of the optical indicatrix. The next order electrooptic phenomenon responsible for the rotations is the fifth-order effect.

Independently of classical measurements based on the light intensity modulation, as an interesting note, we can add that the third-order coefficient *R* may be experimentally determined from measurements of the electric-field-induced change ξ in the azimuth of the fast wave. Considering the light beam propagating, along the X_1 direction, the change in the azimuth is given by

$$tan\,(2\xi) = \frac{3RE}{|g_{11} - g_{12}|}\,. \tag{7}$$

We have checked in our previous experiments that when an immersion liquid is used, it is usually possible to apply to the crystal an electric field of amplitude 10^6 V/m. It is possible to read angles with an accuracy better than 0.5 degree. Thus, Eq. (6) shows that, on the basis of Eq. (7), it would be relatively easy to measure the third-order electrooptic effect larger than 10^{-28} m^3V^{-3}, provided that the quadratic electrooptic coefficient of the crystal is known and comparable to that found in the KH_2PO_4 family crystals, i.e. of the order of magnitude 10^{-20}-10^{-21} m^2V^{-2} [8]. It is interesting to note that for expected small magnitudes of ξ, its changes may be observed on the fundamental frequency of the modulating field, not on its third-harmonic.

4. CONCLUSIONS

The 432 symmetry crystals seem to be very promising in attempts to measure the third- and fourth-order electrooptic effects. Unfortunately, compounds of such symmetry are relatively rare.

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ANALIZA MOŻLIWOŚCI POMIARU NIELINIOWYCH EFEKTÓW ELEKTROOPTYCZNYCH W KRYSZTAŁACH O SYMETRII 432

Streszczenie

Pokazano, że mimo eliminacji przez symetrię liniowego efektu elektrooptycznego, w kryształach o symetrii 432 istnieją konfiguracje, w których można spodziewać się wystąpienia efektu trzeciego rzędu. Otrzymane wyniki wskazują, że kryształy o takiej symetrii są obiecującymi materiałami dla pomiarów nieliniowych efektów elektrooptycznych. Przedstawiono makroskopowy opis nieliniowych efektów elektrooptycznych w kryształach o symetrii 432 łącznie z zestawieniem postaci macierzy tensorów opisujących efekty elektrooptyczne do piątego rzędu włącznie.