

**WŁODZIMIERZ KUCHARCZYK, PIOTR GÓRSKI
RAFAŁ LEDZION**

Institute of Physics, Lodz University of Technology, Wólczajska 219,
93-005 Łódź, Poland. e-mail: wlodzimierz.kucharczyk@p.lodz.pl

MATRICES OF THE TENSOR WITH THE INTERNAL SYMMETRY $[V^2][V^5]$

Matrices of the tensor of the internal symmetry $[V^2][V^5]$ are presented for those point symmetry classes for which the matrices have not been published yet. Conditions allowing for measurements of nonlinear electrooptic effects are discussed.

Keywords: nonlinear electrooptic effects, tensor matrices, the tensor of the internal symmetry $[V^2][V^5]$.

1. INTRODUCTION

Up to now, experimental results related to the nonlinear dependence of the electric susceptibility or impermeability on the low-frequency electric field have been discussed in terms of nonlinear electrooptic effects even of the sixth-order (see, e.g. [1]). The matrices of tensors representing nonlinear electrooptic effects up to the fourth-order are known [2]. However, the matrices of the tensor of the internal symmetry $[V^2][V^5]$ related to the fifth-order electrooptic effect have been published only for 12 of all 21 symmetry classes allowing for the effect, i.e. not possessing the inversion symmetry [3, 4].

The aim of this work is to present matrices obtained for the $[V^2][V^5]$ internal symmetry tensor for those symmetry classes for which their form has not been published yet.

2. MATRICES OF THE TENSOR OF SYMMETRY $[V^2][V^5]$

Traditionally, electrooptic coefficients are defined as terms in a power-series expansion in the low-frequency electric field \mathbf{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. [5-7]). The tensor $K_{ijk_1\dots k_f}$ that represents the electrooptic effect of the order f is given by

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

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

It is known that the symmetry of any physical property of a crystal must include the symmetry elements of the point group of the crystal [8]. 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. A well known example is the finding that no physical property that is represented by an odd rank tensor may appear in a centrosymmetric crystal. This means that all components of the tensor are equal to zero.

In this work the shortened, widely used, matrix notation is applied for the symmetrical tensor of the second rank [8], namely;

$$\text{where:} \quad \begin{array}{l} ij \rightarrow \mu, \\ 11 \rightarrow 1, 22 \rightarrow 2, 33 \rightarrow 3, 23 \rightarrow 4, 13 \rightarrow 5, 12 \rightarrow 6 \end{array} \quad (2)$$

Thus, the shortened matrix notations for the electrooptic tensor of the fifth-order is,

$$L_{ijklmno} = L_{\mu\kappa\lambda mno}.$$

As previously [2-4], to obtain the matrices of the tensor $L_{\mu\kappa\lambda mno}$, 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 [9], for the point symmetry classes listed in Table 1, the functions describing the dependence of the impermeability tensor up to the fifth power of the electrical field were 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 [10, 11]. These numbers were computed using the formula

$$n = \frac{1}{N} \sum_{g \in F} \chi^0(g), \quad (3)$$

where F is the point group of a crystal containing symmetry elements g , N is the order of the group F and $\chi^0(g)$ is the character of corresponding matrix transformation.

The results obtained for those crystal classes for which the form of the tensor of the internal symmetry $[V^2][V^5]$ has not been published yet are gathered in Table 1.

3. DISCUSSION

Usually, to relate the electrooptic properties of the paraelectric and ferroelectric phases intrinsic properties defined, analogously to the tensor $K_{ijk_1 \dots k_f}$, however, now in terms of the induced polarization \mathbf{P} instead of the modulating electric field, are employed, namely;

$$M_{ijk_1 \dots k_f} = \frac{1}{f!} \left(\frac{\partial^f \eta_{ij}(\omega)}{\partial P_{k_1} \dots \partial P_{k_f}} \right) . \quad (4)$$

According to their definitions the coefficients $K_{ijk_1 \dots k_f}$ and $M_{ijk_1 \dots k_f}$ are connected by the expression

$$K_{ijk_1 \dots k_f} = M_{ijk_1 \dots k_f} \varepsilon_0^f (\varepsilon_{k_1} - 1)(\varepsilon_{k_2} - 1) \dots (\varepsilon_{k_f} - 1), \quad (5)$$

where ε_0 is the permittivity of free space. In temperatures close to the phase transition the electric permittivity increases significantly, sometimes even several orders of magnitude. The intrinsic coefficients are widely assumed to have a negligible dependence on temperature and are roughly equal both in the paraelectric and ferroelectric phases (see, e.g. [12]). Thus, investigations in the vicinity of the phase transition temperature seems to be a promising method of measuring the nonlinear electrooptic effects. Furthermore, it is known that the spontaneous polarization in ferroelectrics reaches very large magnitudes. Thus, nonlinear electrooptic effects may also contribute to the spontaneous birefringence appearing in the polarized phase. As shown previously, (see, e.g. [13, 14]), in these measurements, the effect of errors in cutting and alignment of the investigated crystal sample on the nonlinear response has to be considered.

Table 1. The form of the tensor $L_{\mu\kappa\lambda\nu\sigma}$ of the internal symmetry $[V^2][V^5]$, the shortened matrix notation given in Eq. (2) is used

The crystal symmetry system: the symmetry class, (the number of non-zero and independent components, respectively)	The components of the tensor
Triclinic system: class 1 (126, 126)	111111, 122222, 133333, 111112, 111113, 111122, 111123, 111133, 111222, 111223, 111233, 111333, 112222, 112223, 112233, 112333, 113333, 122223, 122233, 122333, 123333, 211111, 222222, 233333, 211112, 211113, 211122, 211123, 211133, 211222, 211223, 211233, 211333, 212222, 212223, 212233, 212333, 213333, 222223, 222233, 222333, 223333, 311111, 322222, 333333, 311112, 311113, 111122, 111123, 311133, 311222, 311223, 311233, 311333, 112222, 112223, 312233, 312333, 313333, 322223, 322233, 122333, 123333, 411111, 422222, 433333, 411112, 411113, 411122, 411123, 411133, 411222, 411223, 411233, 411333, 412222, 412223, 412233, 412333, 413333, 422223, 422233, 422333, 423333, 511111, 522222, 533333, 511112, 511113, 511122, 511123, 511133, 511222, 511223, 511233, 511333, 512222, 512223, 512233, 512333, 513333, 522223, 522233, 522333, 523333, 611111, 622222, 633333, 611112, 611113, 611122, 611123, 611133, 611222, 611223, 611233, 611333, 612222, 612223, 612233, 612333, 613333, 622223, 622233, 622333, 623333,
Tetragonal system: class $\bar{4}$ (30, 57)	133333=-233333, 111113=-222223, 111123=212223, 111223=-211223, 111333=-222333, 112223=211123, 112333=212333, 122223=-211113, 122333=-211333, 311113=-322223, 311123=312223, 311333=-322333, 312333, 411111=522222, 422222=-511111, 411112=-512222, 411122=511222, 411133=522233, 411222=-511122, 411222=-512233, 412222=511112, 412233=511233, 413333=523333, 422233=-511133, 423333=-513333, 633333, 611113=622223, 611123=-612223, 611223, 611333=622333
Tetragonal system: class 422 (12, 24)	111123=-211123, 112223=-212223, 112333=-212333, 311123=-312223, 411111=-522222, 411122=-511222, 411133=-522233, 412222=-511112, 412233=-511233, 413333=-523333, 611113=-622223, 613333=622233

Table 1. (continued)

The crystal symmetry system: the symmetry class, (the number of non-zero and independent components, respectively)	The components of the tensor
<p>Trigonal system: class 3</p> <p>(42, 120)</p>	<p>133333=233333, 122333=211333=111333-612333, 111333=222333, 112333=-212333=-2•611333=2•622333, 113333=-213333=-623333, 123333=-223333=613333, 6•311111=-3•311122=-2•312222, 333333, 2•311112=3•311222=-6•322222, 3•311133=-312233, 2•311113=311223=2•322223, 3•311133=-312233, 311233=-3•322233, 311333=322333, 411133=412233=-511233=-522233, 411233=422233=511133=512233, 2•411333=-2•422333=512333, 413333=-523333, 412333=-2•511333=2•522333, 423333=513333, 111111=A, 211111=B, 622222=C, 111122=-3A+B-6C, 112222=-3A+2C, 211122=A-3B+6C, 212222=-3A-2C, 611112=A-B+C, 611222=-3A+3B-4C, 122222=D, 222222=E, 611111=F, 111112=-3E-2F, 111222=-3D+E+6F, 211112=-3D+2F, 211222=D-3E-6F, 511122=3(D-E), 612222=-D+E+3F, 111113=G, 122223=H, 222223=I, 111223=-3G+H+4I, 211113=-G+H+I, 211223=3G+H-2I, 611123=-G-H+2I, 612223=3G-H-2I, 111133=J, 211133=K, 112233=J-2K, 212233=-2J-K, 611233=622233=0.5(-J+K), 122233=L, 222233=M, 111233=-L-2M, 211233=-2L-M, 611133=612233=0.5(L-M), 411111=N, 522222=P, 411122=-4•N-6•P, 412222=3•N+2•P, 511112=2N+3P, 511222=-6N-4P, 422222=Q, 511111=R, 411112=3Q-2R, 411222=-4Q+6R, 511122=6Q-4R, 512222=-2Q+3R, 411113=S, 422223=T, 411223=-3(S+T), 511123=-S-3T, 512223=3S+T, 511113=U, 522223=V, 411123=U+3V, 412223=-3U-V, 511223=-3(U+V), 611113=W, 622223=Y, 111123=-211123=-W-3Y, 112223=-212223=3W+Y, 611223=-3(W+Y)</p>

Table 1. (continued)

The crystal symmetry system: the symmetry class, (the number of non-zero and independent components, respectively)	The components of the tensor
Trigonal system: class 32 (18, 54)	$112333=-212333=-2\cdot611333=2\cdot622333,$ $112222=-213333=-623333, 6\cdot311111=-3\cdot311122=-2\cdot312222,$ $3\cdot311133=-312233, 411133=412233=-511233=-522233,$ $412333=-2\cdot511333=2\cdot522333, 413333=-523333,$ $111111=A, 211111=B, 622222=C,$ $111122=-3A+B-6C, 112222=-3B+2C, 211122=A-3B+6C,$ $212222=-3A+2C, 611112=A-B+3C, 611222=-3A+3B-4C,$ $111133=D, 211133=E,$ $112233=-D-2E, 212233=-2D-E, 2\cdot611233=2\cdot622233=-D+E,$ $411111=F, 522222=G,$ $411122=-6F-4G, 412222=2F+3G, 511112=3F+2G,$ $511122=-4F-6G,$ $511113=H, 522223=I,$ $411123=H+3I, 412223=-3H-I, 511223=-3(H+I),$ $611113=J, 622223=K,$ $111123=-211123=-J-3K, 112223=-212223=3J+K,$ $611223=-3(J+K)$
Hexagonal system: class $\bar{6}$ (22, 61)	$113333=-213333=-623333, 123333=-223333=613333,$ $6\cdot311111=-312233, 311233=-3\cdot322233,$ $2\cdot411333=-2\cdot422333=512333, 4123444=-2\cdot511333=2\cdot522333,$ $111111=A, 211111=B, 622222=C,$ $111122=-3A+B-6C, 112222=-3B+2C, 211122=A-3B+6C,$ $611222=-3A+3B-4C, 611112=A-B+3C,$ $122222=D, 222222=E, 611111=F,$ $111112=-3E-2F, 111222=-3D+E+6F, 211112=-3D+2F,$ $211222=D-3E-6F, 611122=(D-E), 612222=-D+E+3F,$ $111133=G, 211133=H,$ $212233=-2G-H, 2\cdot611233=2\cdot622233=-G+H,$ $122233=I, 222233=J,$ $111233=-I-2J, 211233=-2I-J, 2\cdot611133=2\cdot612233=I-J,$ $411113=K, 422223=L,$ $411223=-3(K+L), 511123=-K-3L, 512223=3K+L,$ $511113=M, 522223=N,$ $411123=M+3N, 412223=-3M-N, 511223=-3(M+N)$

Table 1. (continued)

The crystal symmetry system: the symmetry class, (the number of non-zero and independent components, respectively)	The components of the tensor
Hexagonal system: class 6 (20, 56)	$133333=233333, 111333=222333,$ $122333=211333=111333-612333,$ $112333=-212333=-2 \cdot 611333=2 \cdot 622333,$ $333333, 2 \cdot 311113=311223=2 \cdot 322223, 311333=322333,$ $411133=412233=-511233=-522233,$ $411233=422233=511133=512233, 413333=-522233,$ $423333=513333,$ $111113=A, 122223=B, 222223=C,$ $111223=-3A+B+2C, 211113=-A+B+C, 211223=3A+B-2C,$ $611123=-A-B+2C, 612223=3A-B-2C,$ $411111=D, 522222=E,$ $411122=-4D-6E, 412222=3D+2E, 511112=2D+3E,$ $511222=-6D-4E,$ $422222=F, 511111=G,$ $411112=3F-2G, 411223=-4F+6G, 511122=6F-4G,$ $512222=-2F+3G,$ $622222=J, 611113=H,$ $111123=-211123=H+3J, 112223=-212223=-3H-J,$ $611223=-3(H+J)$
Hexagonal system: class $\bar{6}m2$ (11, 31)	$123333=-223333=613333, 2 \cdot 311112=3 \cdot 311222=-6 \cdot 322222,$ $311233=-3 \cdot 322233, 2 \cdot 411333=-2 \cdot 422333=512333,$ $122222=A, 222222=B, 611111=C,$ $111112=-3B-2C, 111222=-3A+B+6C, 211112=-3A+2C,$ $211222=A-3B-6C, 611122=3(A-B), 612222=-A+B+3C,$ $122233=D, 222233=E,$ $111233=-D-2E, 211233=-2D-E, 2 \cdot 611133=2 \cdot 612233=D-E,$ $411113=F, 422223=G, 411223=-3(F+G), 511123=-F-3G,$ $512223=3F+G$
Hexagonal system: class 622 (7, 23)	$112333=-212333=-2 \cdot 611333=2 \cdot 622333,$ $411133=412233=-511233=-522233, 413333=-523333,$ $411111=A, 522222=B,$ $411122=-6A-4B, 412222=2A+3B, 511112=3A+2B,$ $511222=-4A-6B,$ $611113=C, 622223=D,$ $111123=-211123=-C-3D, 112223=-212223=3C+D,$ $611223=-(C+D)$

4. CONCLUSIONS

In our opinion, there are two types of experimental conditions, when an observation of nonlinear electrooptic effects is possible. The first of them is when measurements are performed in crystals in the vicinity of the paraelectric-ferroelectric phase transition temperature. The second one are investigations of the relationship between the spontaneous birefringence and the spontaneous polarization in the ferroelectric phase of crystals.

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MACIERZE TENSORA O SYMETRII WEWNĘTRZNEJ [V²][V⁵]

Streszczenie

Przedstawiono macierze o symetrii wewnętrznej [V²][V⁵] dla tych 12 nieposiadających środka symetrii klas symetrii, dla których macierze te nie były dotychczas publikowane. Przeanalizowano warunki zwiększające szanse obserwacji nieliniowych efektów elektrooptycznych.