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MEASUREMENT OF ELEMENTS OF THE MATRIX OF PIEZOELECTRIC COEFFICIENTS IN A FERROELECTRIC SINGLE CRYSTAL

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Abstract

The paper presents a procedure of measurement of piezoelectric coefficients in a ferroelectric single crystal, on the example of $Gd_2(MoO_4)_3$, using an X-ray diffractometer.

POMIAR ELEMENTÓW MACIERZY WSPÓŁCZYNNIKÓW PIEZOELEKTRYCZNYCH MONOKRYSZTAŁU FERROELEKTRYCZNEGO

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Streszczenie

Przedstawiono procedurę pomiaru, z wykorzystaniem dyfraktometru rentgenowskiego, współczynników piezoelektrycznych monokryształu ferroelektrycznego na przykładzie molibdenku gadolinu $Gd_2(MoO_4)_3$.

Introduction

Piezoelectric coefficients, also referred to as piezoelectric moduli, are among the most important quantities characterizing all piezoelectric crystals, including ferroelectric ones (RUDIAK 1982). This is connected with the fact that the matrixes of these coefficients are present in constitutive equations describing direct and converse piezoelectric effects. At the same time, the values of the above coefficients are the same for both these effects. This indicates that the direct or converse piezoelectric effect can be used for measuring matrix elements. The present paper describes measurement of these coefficients based on a converse piezoelectric effect involving piezoelectric crystal strain caused by an electric field.

Measurements

The matrix form of a constitutive equation describing the converse piezoelectric effect is as follows (RUDIAK 1982):

$$\begin{bmatrix} \varepsilon_1 \\ \varepsilon_2 \\ \varepsilon_3 \\ \varepsilon_4 \\ \varepsilon_5 \\ \varepsilon_6 \end{bmatrix} = \begin{bmatrix} d_{11} & d_{12} & d_{13} \\ d_{21} & d_{22} & d_{23} \\ d_{31} & d_{32} & d_{33} \\ d_{41} & d_{42} & d_{43} \\ d_{51} & d_{52} & d_{53} \\ d_{61} & d_{62} & d_{63} \end{bmatrix} \begin{bmatrix} E_1 \\ E_2 \\ E_3 \end{bmatrix} \tag{1a}$$

This equation may be also presented in an abbreviated form:

$$[\varepsilon_i]_{6\cdot 1} = [d_{ij}]_{6\cdot 3}\,[E_j]_{3\cdot 1} \quad (i=1,2,....,6;\ j=1,2,3) \tag{1b}$$

In the above equation, apart from the matrix of piezoelectric coefficients $[d_{ij}]_{6\cdot 3}$, there are also the strain matrix $[\varepsilon_i]_{6\cdot 1}$ and the electric field matrix $[E_j]_{3\cdot 1}$. The strain matrix is composed of elements of the strain tensor. In its general form, this tensor contains nine elements $\varepsilon_{ij}\ (i,j=1,2,3)$. However, if there are no torsional moments in a single crystal, the following equalities are fulfilled:

$$\varepsilon_{12} = \varepsilon_{21},\ \ \varepsilon_{13} = \varepsilon_{31},\ \ \varepsilon_{23} = \varepsilon_{32} \tag{2}$$

and the number of tensor elements is reduced to six. The following dependencies can be observed between these elements and elements of the strain matrix from equation (1a):

$$\begin{aligned} &\varepsilon_1 = \varepsilon_{11}\ , \quad \varepsilon_2 = \varepsilon_{22}\ , \quad \varepsilon_3 = \varepsilon_{33} \\ &\varepsilon_4 = 2\varepsilon_{23}\ , \quad \varepsilon_5 = 2\varepsilon_{13}\ , \quad \varepsilon_6 = 2\varepsilon_{12} \end{aligned} \tag{3}$$

The above dependencies result from the following tensor equation (BRECZKO 1986):

$$\varepsilon_n = \varepsilon_{ij} n_i n_j \qquad (i, j = 1,2,3) \tag{4}$$

used for determining strain in any direction, defined in relation to the reference system concerning the single crystal examined. The orientation of this direction is determined by the direction cosines n_i and n_j, i.e. by the cosines of the angles between a given direction and the coordinate axes i i j, forming a coordinate system relating to the single crystal analyzed.

It should be noted that if equations (4) form a system of nine equations, this system enables to determine all nine elements ε_{ij}, $(i, j = 1,2,3)$ of the strain tensor. However, while formulating these equations it is important to measure strains of a given crystal for nine independent directions, as this allows to obtain a linearly independent system. If equations (2) are fulfilled for a given single crystal, the system of nine linearly independent equations can be reduced to the following system of six equations:

$$\begin{cases} \varepsilon_{n(1)} = \varepsilon_{11} n_{1(1)}^2 + \varepsilon_{22} n_{2(1)}^2 + \varepsilon_{33} n_{3(1)}^2 + 2\varepsilon_{12} n_{1(1)} n_{2(1)} + 2\varepsilon_{13} n_{1(1)} n_{3(1)} + 2\varepsilon_{23} n_{2(1)} n_{3(1)} \\ \varepsilon_{n(2)} = \varepsilon_{11} n_{1(2)}^2 + \varepsilon_{22} n_{2(2)}^2 + \varepsilon_{33} n_{3(2)}^2 + 2\varepsilon_{12} n_{1(2)} n_{2(2)} + 2\varepsilon_{13} n_{1(2)} n_{3(2)} + 2\varepsilon_{23} n_{2(2)} n_{3(2)} \\ \varepsilon_{n(3)} = \varepsilon_{11} n_{1(3)}^2 + \varepsilon_{22} n_{2(3)}^2 + \varepsilon_{33} n_{3(3)}^2 + 2\varepsilon_{12} n_{1(3)} n_{2(3)} + 2\varepsilon_{13} n_{1(3)} n_{3(3)} + 2\varepsilon_{23} n_{2(3)} n_{3(3)} \\ \varepsilon_{n(4)} = \varepsilon_{11} n_{1(4)}^2 + \varepsilon_{22} n_{2(4)}^2 + \varepsilon_{33} n_{3(4)}^2 + 2\varepsilon_{12} n_{1(4)} n_{2(4)} + 2\varepsilon_{13} n_{1(4)} n_{3(4)} + 2\varepsilon_{232} n_{2(4)} n_{3(4)} \\ \varepsilon_{n(5)} = \varepsilon_{11} n_{1(5)}^2 + \varepsilon_{22} n_{2(5)}^2 + \varepsilon_{33} n_{3(5)}^2 + 2\varepsilon_{12} n_{1(5)} n_{2(5)} + 2\varepsilon_{13} n_{1(5)} n_{3(5)} + 2\varepsilon_{232} n_{2(5)} n_{3(5)} \\ \varepsilon_{n(6)} = \varepsilon_{11} n_{1(6)}^2 + \varepsilon_{22} n_{2(6)}^2 + \varepsilon_{33} n_{3(6)}^2 + 2\varepsilon_{12} n_{1(6)} n_{2(6)} + 2\varepsilon_{13} n_{1(6)} n_{3(6)} + 2\varepsilon_{23} n_{2(6)} n_{3(6)} \end{cases} \tag{5}$$

If the above equations are to form a linearly independent system, strain measurements must be taken for six independent directions. The independence of directions means in this case that these directions are not interrelated with the symmetry elements of a single crystal. In the system of equations (5) successive six directions are denoted by consecutive natural numbers given in parentheses. This means that e.g. the symbol $n_{1(1)}$ denotes the cosine of the angle between direction 1 and coordinate axis 1 (x-axis) of the coordinate system, connected with a given sample of a single crystal. Analogically, the symbol $n_{2(3)}$ denotes the cosine of the angle between direction 3 and coordinate axis 2 (y-axis) of the coordinate system, connected with a given sample. It should be stressed that substituting equalities (3) into the system of equations (5), makes it possible to obtain a system

of six equations, enabling to determine six elements of the strain matrix from equation (1a). However, this procedure should be preceded by measurement of strains on the left side of the equality sign in the system of equations (5), and of the values of all direction cosines $n_{i(l)}$; $(i=1,2,3;\ l=1,2,...,6)$ present in this system of equations.

Measurements of the strains $\varepsilon_{n(l)}$, $(l=1,2,...,6)$ were performed using X-ray diffraction, exposing the crystal examined to X-radiation and measuring the Bragg angles, at which diffraction occurred at successive planes of a unit cell of a single crystal of $Gd_2(MoO_4)_3$. The Bragg angles were measured with a goniometer, being a part of a DRON-3 X-ray diffractometer. Simultaneously, a crystal sample was placed in a special holder, to measure the angles necessary for determining direction cosines. The holder enabled sample rotation around two perpendicular axes of revolution, as well as mounting electrodes producing an electric field. The relationship between the Bragg angle and strain in a given direction can be described by the following formula (SENCZYK 1999, BRECZKO 1986):

$$\varepsilon_{n(l)} = -\mathrm{ctg}\,\Theta_{0(l)}\Delta\Theta_{(l)} \quad (l=1,2,...,6) \tag{6}$$

where:

$\Theta_{0(l)}$ – angular position of the center of gravity of a diffraction reflection, without an electric field;

$\Delta\Theta_{(l)}$ – changes in the angular position of the center of gravity of a diffraction reflection, caused by an electric field. These changes should be expressed in radians.

The strains determined on the basis of the above dependence for six locations of a single crystal (l = 1, 2, ..., 6) are included in the system of equations (5). This allows to determine elements of the strain matrix from equation (1a), but this must be preceded by calculating direction cosines present in the above system of equations, and numerical solving of this system. The electric field matrix from equation (1a) can be determined on the basis of sample position in relation to the electrodes producing a given electric field. Thus, the only unknown is the matrix of piezoelectric coefficients. In order to determine 18 unknown elements of this matrix, the sample position in relation to electrodes producing an electric field has to be changed twice. The measuring procedure is the same as described above. In this way two equations analogical to equation (1) are obtained. The matrix of piezoelectric coefficients present in these equations is the same as in equation (1a); only the strain matrix and electric field matrix are different. Three equations formulated in this way may be summarized as the following matrix equation:

$$\begin{bmatrix} \varepsilon_1^1 & \varepsilon_1^2 & \varepsilon_1^3 \\ \varepsilon_2^1 & \varepsilon_2^2 & \varepsilon_2^3 \\ \varepsilon_3^1 & \varepsilon_3^2 & \varepsilon_3^3 \\ \varepsilon_4^1 & \varepsilon_4^2 & \varepsilon_4^3 \\ \varepsilon_5^1 & \varepsilon_5^2 & \varepsilon_5^3 \\ \varepsilon_6^1 & \varepsilon_6^2 & \varepsilon_6^3 \end{bmatrix} = \begin{bmatrix} d_{11} & d_{12} & d_{13} \\ d_{21} & d_{22} & d_{23} \\ d_{31} & d_{32} & d_{33} \\ d_{41} & d_{42} & d_{43} \\ d_{51} & d_{52} & d_{53} \\ d_{61} & d_{62} & d_{63} \end{bmatrix} \begin{bmatrix} E_1^1 & E_1^2 & E_1^3 \\ E_2^1 & E_2^2 & E_2^3 \\ E_3^1 & E_3^2 & E_3^3 \end{bmatrix} \tag{7}$$

The superscripts at elements of the strain matrix and electric field matrix denote positions of the single crystal analyzed in relation to electrodes producing an electric field. The measuring procedure described in this paper shows that the unknown matrix from equation (7) is the matrix of piezoelectric coefficients, which can be determined by pre- and postmultiplication of equation (7) by a matrix inverse to the electric field matrix. Then the following matrix equation is obtained:

$$\begin{bmatrix} d_{11} & d_{12} & d_{13} \\ d_{21} & d_{22} & d_{23} \\ d_{31} & d_{32} & d_{33} \\ d_{41} & d_{42} & d_{43} \\ d_{51} & d_{52} & d_{53} \\ d_{61} & d_{62} & d_{63} \end{bmatrix} = \begin{bmatrix} \varepsilon_1^1 & \varepsilon_1^2 & \varepsilon_1^3 \\ \varepsilon_2^1 & \varepsilon_2^2 & \varepsilon_2^3 \\ \varepsilon_3^1 & \varepsilon_3^2 & \varepsilon_3^3 \\ \varepsilon_4^1 & \varepsilon_4^2 & \varepsilon_4^3 \\ \varepsilon_5^1 & \varepsilon_5^2 & \varepsilon_5^3 \\ \varepsilon_6^1 & \varepsilon_6^2 & \varepsilon_6^3 \end{bmatrix} \begin{bmatrix} E_1^1 & E_1^2 & E_1^3 \\ E_2^1 & E_2^2 & E_2^3 \\ E_3^1 & E_3^2 & E_3^3 \end{bmatrix}^{-1} \tag{8}$$

This equation can be solved by numerical methods, obtaining elements of the matrix of piezoelectric coefficients.

Results of measurements

Table 1 presents the results of measurements of elements of the matrix of piezoelectric coefficients of a single crystal of $Gd_2(MoO_4)_3$ placed in the holder of a goniometer of an X-ray diffractometer. The measurements were performed following the procedure described in this paper. The values included in Table 1 were obtained for electric field intensity of 2 kV/cm. Figure 1 shows an example of a diffraction reflection obtained for a sample exposed to X-radiation, not affected by an electric field. Figure 2 illustrates shows a diffraction reflection obtained for a sample exposed to X-radiation,

Table 1

Elements of the matrix of piezoelectric coefficients

d_{11}	d_{12}	d_{13}	d_{21}	d_{22}	d_{23}	d_{24}	d_{32}	d_{33}
0.3 10^{-9}	0.2 10^{-9}	2.1 10^{-9}	0.3 10^{-9}	0.2 10^{-9}	1.8 10^{-9}	2.3 10^{-9}	0.3 10-9	0.8 10-9

m/V

d_{41}	d_{42}	d_{43}	d_{51}	d_{52}	d_{53}	d_{61}	d_{62}	d_{63}
0.4 10^{-9}	0.7 10^{-9}	0.1 10^{-9}	0.5 10^{-9}	0.03 10^{-9}	1.2 10^{-9}	0.1 10^{-9}	0.05 10^{-9}	0.02 10^{-9}

m/V

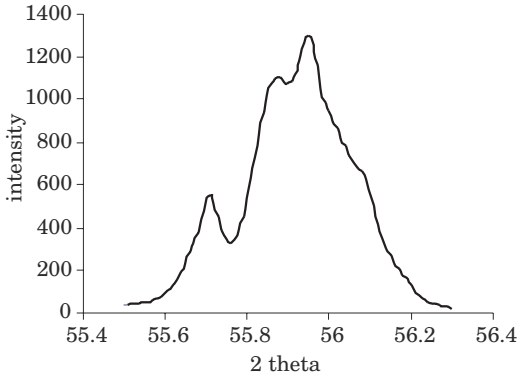

Fig. 1. XRD pattern of a single crystal $Gd_2(MoO_4)_3$ of without an electric field (NiKα)

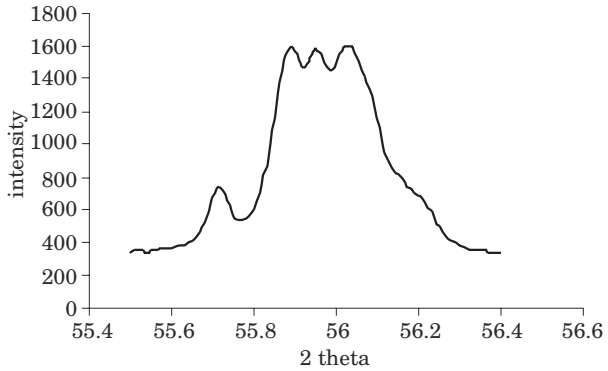

Fig. 2. XRD pattern of a single crystal of $Gd_2(MoO_4)_3$ (NiKα)

affected by an electric field of intensity 2 kV/cm. The first diffraction reflection enables to determine the angle $\Theta_{0(l)}$, present in equation (6), and a comparison of both reflections allows to calculate the angular difference $\Delta\Theta_{(l)}$, also present in equation (6).

Conclusions

The procedure of measurement of elements of the matrix of piezoelectric coefficients with an X-ray diffractometer, described in the paper, is time- and labor-consuming, but its results can be applied in practice. It requires first of all proper mounting of the sample and adjusting the X-ray diffractometer. This should be followed by finding at least six positions of the sample in relation to X-radiation, enabling to obtain a diffraction reflection. The next step is taking 18 diffraction measurements, six for three different positions of the sample in relation to electrodes producing an electric field. Diffraction reflections, direction cosines and elements of the electric field matrix are obtained for each of the above measurements, thus providing the basis for determining elements of the matrix of piezoelectric coefficients.

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